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EVALUATION OF WELDING TECHNIQUES FOR TUNGSTEN - URANIUM DIOXIDE COMPOSITES

by Thomas J. Moore and Gordon K. Watson

Lewis Research Center
Cleveland, Ohio

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EVALUATION OF WELDING TECHNIQUES FOR TUNGSTEN - URANIUM DIOXIDE COMPOSITES (U)

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SUMMARY

The fabrication of nuclear rocket reactor fuel elements from flat tungsten - uranium dioxide composite plates is dependent on the use of suitable welding techniques. Ordinary fusion welding techniques were not applicable to the composites because of uranium dioxide vaporization and the resulting porosity in the weld zone. Therefore, six welding techniques in which fusion of the composites was not involved were investigated. The processes studied were gas-pressure bonding, magnetic force welding, vacuum hot-press bonding, vapor deposition welding, gas tungsten-arc brazing, and a special electron beam welding technique. The resulting joints were evaluated nondestructively and metallographically and by high-temperature (2500° C) tensile tests and elevated-temperature (540° and 650° C) bend tests.

The results of this evaluation indicate that the program goals can be met by gas-pressure bonding at 1650° C and 30 000 psi for 2 hours. At the present state of development, gas-pressure bonding is the only method studied that is capable of producing high integrity weldments in all the required joint configurations. Further development of the brazing and magnetic force welding processes also might lead to their use in this application.

INTRODUCTION

A tungsten - water-moderated nuclear reactor concept has been investigated at the NASA Lewis Research Center for use in a nuclear rocket for space propulsion. The fuel elements in this thermal reactor concept would consist of 10- to 35-volume-percent uranium dioxide (UO_2) particles uniformly dispersed in a continuous tungsten (W) matrix. These fuel elements will be required to operate in rapidly flowing hydrogen (the rocket propellant) at temperatures of about 2500° C for up to 10 hours.

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A powder-metallurgy and hot-rolling technique has been developed at the Lewis Research Center for the fabrication of thin (0.021 in.), flat tungsten - uranium dioxide ($W-UO_2$) plates (ref. 1). In this process, the major surfaces of the fuel plates are clad with a thin (0.001 in.) layer of unfueled tungsten by bonding wrought tungsten foil to the fuel plates. This external cladding is required to minimize the loss of fuel by vaporization at elevated temperatures.

It also was shown in reference 1 that the $W-UO_2$ plates could be hot formed into various shapes, such as cylinders, which could be used as fuel element components. Before useful fuel elements could be fabricated from these plates, however, suitable welding techniques had to be developed for the joining of $W-UO_2$ composites. Conventional fusion welding techniques, such as gas tungsten-arc (TIG) and electron beam welding, were not directly applicable to $W-UO_2$ composites because of excessive porosity in the weld zone as a result of UO_2 vaporization. Therefore, various other welding techniques in which fusion of the composites would not be involved were investigated both under NASA contracts and at the NASA Lewis Research Center. The welding techniques studied are as follows:

Gas-pressure bonding (NASA contract), reference 2
Magnetic force welding (NASA contract), reference 3
Vacuum hot-press bonding (NASA contract), reference 4
Vapor deposition welding (NASA contract), (unpublished data)
Gas tungsten-arc brazing (NASA contract), reference 5
Special electron beam welding techniques (Lewis)

The first four techniques listed are solid-state welding methods. In this report, these methods are defined as processes by which members are bonded by atomic forces without the presence of a liquid phase at any stage of the process. The major objective of the latter two techniques listed was to avoid melting the composites during joining by using a filler material in the joint. In the brazing method, braze alloys selected had remelt temperatures in excess of 2760° C. In the electron beam process, the mating edges of $W-UO_2$ composite plates were clad with tungsten, and then the electron beam process was employed to fusion weld the edge cladding without melting the composites.

The joints resulting from these programs were to meet the following requirements:

(1) The joints should be fully sound and free of cracks or voids.

(2) The joints should be usable between ambient temperature and 2760° C and should be capable of operating for at least 10 hours at maximum temperature with intermittent cycling to ambient temperature.

(3) Unfueled zones produced by the welding process should be as small as possible (preferably less than 0.020 in.) because unfueled regions cause uneven heating of the fuel element.

(4) The mechanical properties of the joint region should be equal to or greater than the mechanical properties of the $W-UO_2$ composite.

(5) Materials added to the joint should be compatible with tungsten, UO_2, and hydrogen and should have a low nuclear cross section.

(6) Contamination of the composite during welding should be prevented.

(7) Loss of UO_2 from the joint should be prevented both during processing and in service.

All the welding programs were conducted in a similar manner on $W-UO_2$ composites containing 20 volume percent UO_2. The initial part of each program (phase I) consisted of a feasibility study to determine if the process was applicable to $W-UO_2$ composites. If feasibility was demonstrated in the initial studies, additional work was performed to optimize the process parameters (phase II). At the conclusion of each program, welded samples of the configurations and quantities shown in figure 1 were submitted to Lewis Research Center for evaluation.

This report describes the results obtained from the Lewis evaluation of the welded $W-UO_2$ composites. Evaluation techniques included nondestructive tests, metallography, high-temperature tensile tests, and elevated-temperature bend tests. The relative merits of each joining process are described herein, and comments are offered in regard to which welding techniques for $W-UO_2$ composites best met the goals of the program.

WELDING TECHNIQUES

The welding techniques used to produce the specimens for this evaluation are briefly summarized in the following paragraphs and are described in detail in references 2 to 5.

Gas-Pressure Bonding

The $W-UO_2$ composites were successfully welded by the gas-pressure bonding technique (ref. 2). This process used a combination of heat and pressure in an autoclave to achieve a solid-state weld between clean surfaces of the composites.

Prior to bonding, the edges of the composites were ground square to provide clean, flat surfaces (10 to 15 μin. rms). The composites then were placed into a molybdenum container, heat treated in vacuum to remove surface impurities (1100° C for 1 hr), and sealed under vacuum by using the electron beam welding process. A schematic of the autoclave and the design of the container and internal components used for the small butt joints are shown in figure 2. The container was designed so that the isostatic pressure first forced the mating surfaces of the joint into intimate contact by the collapse of the end plates (which were thinner than the side plates). Similar container designs were used for the other configurations required in the program. After the bonding cycle, the molybdenum container was removed from the weldments by dissolution in a nitric acid solution.

At the conclusion of the investigation, all the required specimens were submitted to NASA for evaluation. These weldments were bonded at 1650° C and 30 000 psi for 2 hours. The majority were submitted in the as-bonded condition; however, some of the samples which contained tungsten powder at the joint to improve bonding were heat treated at 2040° C for 2 hours in vacuum after bonding.

Magnetic Force Welding

The magnetic force welding technique utilized an electrical-resistance heating effect at the mating surfaces of the joint in combination with a synchronized forging blow delivered by an electromagnetic device to achieve a solid-state weld. Only a fraction of a second was required to achieve a weld with this process. Thus, this technique was ideally suited for welding W-UO_2 composites because vaporization of UO_2 at the joint region should be minimized. The magnetic-force welding process differs from conventional resistance welding processes in the method of generating the pressure at the joint. Conventional machines use either pneumatic or hydraulic force systems exclusively; whereas, in magnetic force welding, a forging blow is generated by an electromagnet coupled to a movable electrode as shown in the schematic in figure 3. Air pressure is utilized only to provide the initial contact between the mating surfaces.

The major efforts of the magnetic force welding program were to achieve complete metallurgical bonding at the joint interface and to demonstrate process reproducibility. Various welding parameters were investigated: these included weld time, weld current, magnet gap (which controlled the electromagnetic force), and initial pneumatic electrode force. The parameters which yielded the best results for flat plate and cylinder butt joints are given in table I. A limited amount of work was devoted toward reducing the upset at the joints by the use of restraining strips.

TABLE I. - MAGNETIC FORCE WELDING PARAMETERS

Parameter	Small butt joints and tensile blanks	Cylinder butt joints
Weld time, msec	5.1	8.2 to 8.4
Weld current, A	23 000	26 000
Magnet gap, in.	7/8	13/16
Initial pneumatic electrode force, lb	250	200

[redacted]

At the completion of the program, small butt joints, tensile blanks, and cylinder butt joints were welded using the parameters in table I and were submitted for evaluation. No attempt was made on these samples to limit the joint upset with restraining strips. No T-joints were attempted because of time limitations on the program.

Vacuum Hot-Press Bonding

Solid-state welding of W-UO_2 composites was attempted with a vacuum hot press (ref. 4). In this study, resistance heating was used to heat the samples to the bonding temperature, and pressure was applied normal to the joint by a small hydraulic press. The bonding operation was performed in a chamber evacuated to 1×10^{-4} torr.

The bonding fixtures for both the small butt and cylindrical butt joints are shown in figure 4. The W-UO_2 composites were separated from the graphite dies and the mandrel by layers of tantalum carbide. Two types of modified scarf joint designs (shown in fig. 4) were used in this program. The single-joint design and an insert-type joint were used for both the small butt joints and the tensile blanks. The more complex insert-type joint was also employed for joining cylinders. The surface preparation for these joints consisted of grinding the surfaces and then cleaning the surfaces ultrasonically in an organic solvent.

The bonding pressures, temperatures, and times were investigated over a wide range, and the sets of preferred bonding procedures were: (1) 2000° C for 2 hours at 5500 psi plus a postbond heat treatment for 2 hours at 2500° C and (2) 2200° C for 2 hours at 5500 psi with no postbonding heat treatment. The surfaces of the samples that were heat treated at 2500° C were clad with tungsten by a vapor-deposition technique (hydrogen reduction of tungsten hexafluoride) after bonding in order to prevent fuel loss during the heat treatment.

At the completion of the investigation, the four required joint configurations were supplied to Lewis for evaluation. These samples were representative of both joint designs and of both bonding temperatures.

Vapor Deposition Welding

The feasibility of butt welding W-UO_2 composites by vapor deposition techniques was investigated by using the hydrogen reduction of tungsten hexachloride (Unpublished data obtained under NASA Contract by Sylvania Electric Products Inc., Chemical and Metallurgical Division). This program was terminated at the conclusion of the initial study (phase I) because feasibility was not demonstrated.

The apparatus and sample holder arrangement used for vapor-deposition welding is shown in figure 5. The samples to be joined were mounted in the graphite holder as shown. Areas where vapor-deposited tungsten was not desired were masked with alumina. The joint and holder were inductively heated to the reaction temperature, and tungsten was allowed to deposit in a V-groove joint.

The tungsten deposit was generated at the joint by the hydrogen reduction of tungsten hexachloride by the reaction

$$WCl_6 + 3H_2 \longrightarrow W + 6HCl$$

The granular tungsten hexachloride was vaporized by heating to 160° C. Pressure in the hydrogen feed system was controlled between 5 and 20 torr. During deposition, the hydrogen and tungsten hexachloride gases reacted at the heated surface (about 1000° C) of the composite plate to produce an atom-by-atom buildup of tungsten at the joint.

Chemical analyses of the joined composites indicated that very little carbon or halide pickup was encountered during welding. The carbon content varied from 8 to 19 parts per million, and chlorine varied from 1 to 2.5 parts per million. These analyses, however, are an average for the entire sample, and it is possible that some localized contamination within the small vapor deposited tungsten joint could have occurred.

Variables investigated included butt joint design, surface preparation, feed ratio of hydrogen and tungsten hexachloride, and deposition temperature. After preliminary experiments with straight butt and single V-joints with included angles from 40° to 90°, a single V-groove design with a 90° included angle was chosen for the bulk of the work. The surface preparation technique that showed the most promise was an ultrasonic etch in Murakami's reagent ($K_3Fe(CN)_6$ + KOH) followed by an ultrasonic rinse in methanol. The optimum mole ratio of the hydrogen to tungsten hexachloride was found to be 33 to 1. Deposition temperatures of 940° to 1050° C were investigated; a temperature of 1015° C gave the best results.

Small butt joints in the $W-UO_2$ composites were prepared by using the optimum parameters listed previously. After welding, the specimens were surface ground to remove the excess vapor deposited tungsten and then heat treated at 1925° C in vacuum for 1 hour. These samples were submitted to Lewis for evaluation.

Gas Tungsten-Arc Brazing

Several special, high-temperature braze alloys were developed for use with the gas tungsten-arc (TIG) process as a heat source. This brazing technique, classified herein as a welding process, is described in detail in reference 5. Gas tungsten-arc heating was chosen over furnace heating principally because the application of heat in the gas

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tungsten-arc process was quite localized and, thus, the heat affected region from which UO_2 might vaporize was minimized.

Eighteen braze alloys with melting points above 2540° C, low nuclear cross sections, and a minimum tendency to hydride were initially investigated. These alloys were prepared in a button furnace in a high purity argon environment by using a nonconsumable tungsten electrode. The buttons then were crushed to coarse particles (-16/+25 mesh) for brazing with a tungsten-arc as illustrated in figure 6. The alloys were evaluated on the basis of melting point, wettability, and the ability to produce sound butt joints with minimum base metal dissolution. The number of alloys was eventually reduced to three. The three alloys selected for the remainder of the study were tungsten - 25 percent osmium, tungsten - 50 percent molybdenum - 3 percent rhenium, and molybdenum - 5 percent osmium (percent by weight).

At the conclusion of the program, W-UO_2 specimens of all configurations except for the T-joints were prepared for evaluation at Lewis. Most of the joints were produced by using the tungsten - 25 percent osmium alloy. The following typical gas tungsten-arc brazing parameters were used in an atmosphere chamber:

(1) Heat source: Automatic gas tungsten-arc process with a pointed 1/16-inch-diameter, tungsten - 2 percent thorium dioxide electrode operated at 8 volts, 80 amperes, direct-current straight polarity; travel speed of the arc, 10 inches per minute

(2) Atmosphere: Argon containing less than 15 parts per million oxygen and 15 parts per million water vapor

(3) Joint: Straight butt with initial gap of 0.010 to 0.012 inch

(4) Edge preparation: UO_2 particles leached from edges with nitric acid

(5) Alloys: Tungsten - 25 percent osmium, tungsten - 50 percent molybdenum - 3 percent rhenium and molybdenum - 5 percent osmium

Special Electron Beam Welding Techniques

The objective of this study, which was conducted at the Lewis Research Center, was to produce a fusion welded joint but, at the same time, avoid fusion of the W-UO_2 composites by utilizing a tungsten cladding on the mating edges of the composite. These unfueled tungsten edges were then fused together by using the electron beam process. A sketch of the joint cross section is shown in figure 7.

The edges of the composites were clad by using the hot-substrate plasma spraying technique developed by Grisaffe and Spitzig (ref. 6). This technique resulted in metallurgical bonding between the plasma sprayed tungsten and the tungsten substrate. After the specimens had been sprayed, they were ground to the dimensions shown in figure 7.

[redacted]

Electron beam welding was accomplished by using a 150-kilovolt, 13.3-milliampere machine. Actual welding parameters employed were 150 kilovolts and 3.3 milliamperes with an inline beam deflection of 0.020-inch at a travel speed of 100 inches per minute. Small butt joints in the W-UO_2 composites were prepared for evaluation by using the optimum parameters; joints in the other configurations were not attempted.

EVALUATION TECHNIQUES

The joints in the W-UO_2 composites were evaluated by various methods including nondestructive inspection, metallographic examination, bend tests, and tensile tests.

Nondestructive Inspection

Both radiographic and dye-penetrant examinations were conducted on all the joints received for evaluation. Any defects detected by radiography were also detected with the dye-penetrant procedures. For most of the programs, therefore, only the dye-penetrant results are presented.

Metallographic Examination

Defect indications were utilized in determining where metallographic sections were to be taken. Minimization of cracking during the metallographic preparation was accomplished by having all cuts in the welded samples made with electrodischarge machining techniques, and by having the weldments mounted in an epoxy resin that was cured at room temperature. The samples were then polished metallographically. Murakami's reagent was employed for all etched samples.

Bend Tests

Bend tests were conducted on 1/4- by 1-inch sections of welded samples by using a commerical hydraulic-driven compression testing machine in order to obtain some information regarding the strength and soundness of the joints. A 4.5 (0.090-in.) bend radius was employed, and the axis of bending was parallel to the joints. The specimens were heated to the desired test temperature (540° or 650° C) with a small resistance wound muffle furnace which surrounded the bend-test fixture. These elevated test tem-

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peratures were employed to avoid the ductile-brittle transition temperature range of the composite. Temperature was monitored with a Chromel-Alumel thermocouple mounted on the punch. The 540° C tests were conducted in air, and the 650° C tests were conducted in flowing argon to prevent oxidation of the tungsten. The bend specimens were soaked at temperature for 5 minutes and then loaded at a crosshead speed of 2 inches per minute until either fracture occurred or a bend angle of at least 90° was achieved. If a 90° bend angle was achieved with no evidence of cracks, it was considered that the ductile-brittle transition temperature was below the test temperature.

Tensile Tests

High-temperature (2500° C) tensile tests were conducted on the welded samples by using the procedure described in reference 7. Each tensile sample was 3/4 inch wide by 6 inches long and had a reduced section of 3/8 inch by $1\frac{1}{4}$ inches. The reduced sections were formed by electrodischarge machining, and the joints were located in the center of the reduced section. Any excess metal at the surfaces of the joints was removed by grinding the joint region to within 0.001 inch of the surface of the samples. The tensile samples were heated in vacuum ($\sim 5\times10^{-5}$ torr) to the test temperature in approximately 30 minutes and then were loaded to failure at a crosshead speed of 0.03 inch per minute.

RESULTS AND DISCUSSION

Gas-Pressure Bonding

Typical gas-pressure bonded samples of the four required configurations are shown in figure 8. All these joints appeared excellent under visual examination. However, the joints were delineated (fig. 8) because of slight surface discontinuities. The results of the more extensive tests performed on these and similar samples are described in the following subsections by the type of configuration evaluated.

Small butt joints. - The dye-penetrant inspection results for 18 small butt joints are presented in table II. Only joint 14 was found to be defective.

All 18 specimens were cross sectioned to metallographically evaluate bond quality and reproducibility. These specimens, with the exception of joint 14, were found to be essentially free of defects, and the joints were completely bonded with excellent clad-to-clad bonding. Photomicrographs of the joints produced by the three procedures used with this welding method are shown in figure 9. Addition of tungsten powder to the joint (fig. 9(b)) did not improve the grain growth across the joint interface. Heat treatment of

[redacted]

TABLE II. - EVALUATION RESULTS FOR GAS-PRESSURE BONDED SMALL BUTT JOINTS

Specimen	Powder at interface	Postbonding heat treatment	Dye-penetrant defect indications	Bend-test results at 650° C, deg
4	No	None	None	90
6				90
16				(a)
17				
21				
22				
8	Yes	None	None	90
10				90
11				(a)
12				
13				
15				
7	Yes	2040° C for 2 hr	None	(a)
9			None	(a)
14			3/8-in. defect at joint	(a)
18			None	90
19			None	90
20			None	(a)

[a]Specimen not tested.

the weldments containing powder additions (fig. 9(c)) was not successful in eliminating the fine grains generally observed at the bond line. There was, however, a tendency for increased grain growth across the bond line. On the basis of metallographic examination of all of the small butt joints, no clear cut preference between the three bonding procedures could be determined.

Metallographic examination of the only defective area found on joint 14 indicated a sharp notch-like defect with porosity and small UO_2 particles along the bond line. The reason for the occurrence of this defect is not known.

Bend tests were performed at 650° C on several of the small butt joints in an attempt to determine which bonding procedure yielded the best results. Each of the six specimens indicated in table II was cut, transverse to the joint, into three 1/4- by 1-inch samples. The center piece and one end piece of each weldment were bent with one major surface in tension for the center piece and the opposite surface in tension for the end piece. The other end piece was used for metallography.

TABLE III. - EVALUATION RESULTS [redacted]R GAS-PRESSURE

BONDED TENSILE BLANKS

Specimen	Powder at interface	Postbonding heat treatment	Dye-penetrant defect indications	Ultimate tensile strength at 2500° C, psi
1	No	None	1/16-in. defect at joint	----
3	No	None	None	4025
9	No	None	None	3570
2	Yes	None	Full length of joint	----
10			None	3720
11			Full length of joint	----
12			Slight indication	----
13			None	4000
4	Yes	2040° C for 2 hr	1/4-in. defect at joint	----
5			1/2-in. defect at joint	----
6			None	3640
8			None	3840
Composite base material				3400 to 4400

As indicated in table II, all specimens tested completed a 90° bend. Examination of the samples after bending revealed no evidence of failure in any of the bend tests. These tests showed that all the bonding procedures employed produced high integrity joints, and thus no preference in bonding procedure could be established from this bend-test program.

Tensile blanks. - The results of dye-penetrant examination of 12 tensile blank samples indicated that half the samples had defects (see table III). Apparently some additional work is required on this configuration in order to produce consistently sound joints.

Tensile tests were conducted on the defect-free joints. These samples exhibited tensile strengths (table III) at 2500° C comparable to the tensile strength of the $W-UO_2$ base metal at the same temperature (ref. 7). The tensile fractures of all the gas-pressure bonded specimens, however, occurred at the bond line with very little deformation. On the basis of the tensile data, the effect of powder additions to the joints or postbonding heat treatment is inconclusive.

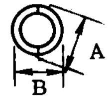
Cylinder butt joints. - Two types of cylinder seam butt designs were submitted. The "full" cylinder design employed only a single longitudinal seam, and the other design used two half cylinders that were bonded along two longitudinal seams. The results

TABLE IV. - EVALUATION RESULTS FOR

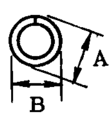
GAS-PRESSURE BONDED CYLINDERS

Specimen	Powder at interface	Type	Dimension, in.		Dye-penentrant defect indications
			A	B	
5	No	Halves[a]	0.552	0.552	Five small flaws at one joint on i.d.
2	Yes		.552	.552	Four small flaws at one joint on i.d.; one small flow on o.d.
3			.552	.552	1/4- to 1/2-in. defects at joints on i.d. and o.d.; two 1/16-in. transverse cracks on o.d. at one joint
4			.553	.552	1/4-in. defect on o.d. at one joint
6			.550	.549	1/4-in. defect at o.d. and i.d. of one joint
8		Full[b]	.531	.531	None
9		Full	.531	.531	None

[a]Cylinder made from two half cylinders.



[b]Cylinder made from one piece.



of dye-penetrant examination of the W-UO_2 cylinders are given in table IV. On the basis of this examination, it appears that the full cylinder joint design yielded better results.

Included in table IV are the results of diametral measurements on the gas-pressure bonded cylinders. The diameters were measured at two different locations on the circumference of each cylinder as indicated. All the cylinders were within 0.001 inch of being circular. These measurements however, were not taken at the joints where discontinuities may be present. Such joint misalinement can sometimes develop when premature welding occurs as a result of container deficiencies. This localized misalinement, which was as much as 0.002 inch, is probably responsible for many of the defects observed in the cylinders. It was confined to the immediate vicinity of the joints, however, because the cylinder was pressed on a mandrel.

Each of the cylinders was cross sectioned for metallographic examination. The cylinders which exhibited defect indications by dye-penetrant examination were sectioned at these locations. Figure 10(a) shows the cross section of a joint in a cylinder fabricated from a full cylinder. A joint obtained by using two half cylinders is shown in figure 10(b). Examination of the cross sections indicated that the joint alinement problems ap-

[redacted]

[redacted]

pear to be more severe when half cylinders were utilized (fig. 10(b)) than when full cylinders were bonded along a single seam (fig. 10(a)).

Grain growth across the bond line of the joints in the half cylinders was quite limited (fig. 10(b)), but for the full cylinders, small grains grew across the bond line (fig. 10(a)).

T-joints. - The six T-joints evaluated were in the as-bonded condition. Tungsten powder was added to all the T-joints prior to bonding. Dye-penetrant examination indicated no defects in four of the joints and small defects in the other two specimens.

Metallographic examination of the sound specimens confirmed the fact that complete bonding was achieved with no significant distortion of the members. Small grains were in evidence at the bond line, and some grain growth occurred across the joint interface for all four specimens. A cross section in the vicinity of a dye-penetrant defect indication in one of the other T-joint specimens is shown in figure 11. This defect, which is just outside the joint, is apparently associated with the loss of a UO_2 particle. The joint itself was sound and well bonded. Based on these inspection results, it is thought that T-joints in W-UO_2 composites can be readily fabricated by gas-pressure bonding.

Discussion. - This study has demonstrated that W-UO_2 composites can be welded by gas-pressure bonding at 1650° C and 30 000 psi for 2 hours. Defect-free, perfectly alined joints were readily produced in the small butt specimens, but some defects were observed in the tensile specimens, the cylinders, and the T-joints. The joint alinement problem encountered in the bonding of half cylinders could possibly be minimized by changes in the container design and/or changes in the joint design from a straight butt to some other joint design, such as a chevron. The joints produced by the full-cylinder technique were free of defects, and joint alinement was acceptable.

The results from both bend tests and tensile tests were inconclusive in determining the best bonding procedure. No differences could be detected between the joints bonded with or without tungsten powder additions to the joints. Heat treating (2040° C for 2 hr) of the joints containing tungsten powder had no apparent effect on mechanical properties. This heat treatment, however, tended to promote grain growth across the bond line.

Tensile strengths of all the weldments were comparable to that of the W-UO_2 base material. All the tensile failures, however, occurred along the original bond line with almost no elongation. Explanations for this mode of failure include a possible concentration of microporosity along the original bond line and the presence of a large number of grain boundaries along the interface.

[redacted]

Magnetic Force Welding

Small butt joints. - Dye-penetrant examination of the 18 small butt joint samples indicated that the joint interfaces of all the samples were sound but that all the samples were cracked transverse to the weld. At least two transverse cracks were detected in each sample, and some samples contained as many as six cracks. The cracks were about 1/8 inch long and extended through the welds, which indicated that the samples probably cracked after the joint was achieved. The cracking problem could possibly be minimized or eliminated by preheating and postheating the joints or by employing low-restraint fixtures. These approaches, however, were not investigated in this study.

Metallography of the welded samples showed that excellent bonding was achieved across the joint interface. The interface was characterized by small tungsten grains and deformed UO_2 particles. A typical joint cross section is shown in figure 12. The black areas are probably regions where UO_2 particles were pulled out during metallographic preparation. The upset in this particular joint (fig. 12) was about 0.010 inch per side, and no clad-to-clad bonding was achieved; however, other joints produced by the contractor (but not submitted for this evaluation study) employed graphite restraining strips which reduced the joint upset to about 0.002 inch per side.

Figure 13 shows the microstructure of a sample cross sectioned through the joint parallel to the major surfaces of the sample. A crack transverse to the joint can be plainly seen. This type of crack was typical of those found in all the magnetic force welded samples.

Bend tests were conducted at 650° C on four of the weldments. The upset at the joints was ground from these weldments prior to testing. All these weldments failed through the weld during bending with essentially no deformation.

Tensile blanks. - Dye-penetrant examination of the tensile blanks indicated that transverse cracks were present in all the joints. Because the tensile load was applied to the samples parallel to the cracks (perpendicular to the joint), it was thought that the cracks would have only a limited effect on tensile results. The tensile results are given in table V. All the specimens fractured in the base metal and not at the weld joint. The strengths obtained were approximately 1100 psi below the average of that obtained previously on the base metal.

Cylinder butt joints. - Transverse cracks, similar to those described previously, were detected by dye-penetrant inspection in all the cylinder butt joints. The joint interfaces, however, appeared to be sound. Fine hairline cracks in a random pattern were detected on the surface of the cylinders about 1/8 inch from the joints. These cracks are thought to be due to the clamping mechanism used to hold the cylinders in the welding fixture and the fact that the base material had very poor ductility at room temperature.

[redacted]

TABLE V. - TENSILE STRENGTH OF MAGNETIC FORCE WELDED TUNGSTEN - URANIUM DIOXIDE COMPOSITES TESTED AT 2500° C

Specimen	Ultimate tensile strength, psi	Fracture location
125-2	2980	Base material
126	2890	Base material
127	2700	Base material
Composite base material	3400 to 4400	-------------

The alinement of the joints appeared satisfactory, and the joint interface was very well bonded as shown in figure 14. This joint is typical of those obtained in the cylinders.

Discussion. - The study on magnetic force welding indicated that this technique offers great potential since solid-state welding can be achieved in W-UO_2 composites in a fraction of a second. Cracking of the samples transverse to the joint, however, was a major problem which was not solved. Thus, before magnetic force welding is applicable to actual joint geometries, additional development work must be done to solve the cracking problem.

The poor ductility observed in the bend tests of the weldments is thought to be the result of the upset at the joints. In the photomicrograph shown in figure 12, it can be seen that the deformed UO_2 particles at the joint tend to be oriented perpendicular to the major surfaces of the weldment. During bending, these particles can act as stress concentrators and thus cause premature failure.

The tensile specimens failed in the base metal rather than through the joint during high-temperature tensile testing. At high temperatures the W-UO_2 composites apparently were not notch sensitive and premature failure of the joints did not occur. The low strengths observed in the base material of these tensile specimens, when compared with the results obtained previously on unwelded specimens of the same type, are not fully understood. One possible explanation is that the restraint produced by the weld tooling introduced small undetected cracks in the specimens during joining.

Vacuum Hot-Press Bonding

Nondestructive inspection of all the vacuum hot-press bonded samples was ineffective because of the complex joint designs and the fact that after bonding most of the samples were clad with tungsten by vapor deposition techniques. Both radiography and dye-

[redacted]

penetrant examination yielded very little useful information. Therefore, all the joints were examined and evaluated metallographically. The following terms were used to describe the condition at the bond line:

Poor - less then 50-percent grain growth across interface (fig. 15(a))

Fair - moderate grain growth across interface (~50 to 75 percent of the area), but small, recrystallized grains still present (fig. 15(b))

Good - more grain growth across interface (>75 percent of the area), but small, recrystallized grains still present (see fig. 15(c))

Excellent - interface not visible, grain growth complete, and grains at the joint indistinguishable from those in the bulk material (fig. 15(d))

Small butt joints. - The bonding parameters and metallographic results for the small butt joints are listed in table VI. The metallographic study indicated that generally

TABLE VI. - METALLOGRAPHIC OBSERVATIONS ON VACUUM

HOT-PRESS BONDED SMALL BUTT JOINTS

Specimen	Insert	Bonding temperature,[a] °C	Tungsten coated	Postbonding thermal treatment[b]	Condition at bond line
43	No	2000	Yes	Yes	Fair
39	↓	↓	↓	↓	Poor
42	↓	↓	↓	↓	Good
41A	↓	↓	↓	↓	Good
30	No	2200	No	No	Good
31	↓	↓	↓	↓	Good
44	↓	↓	↓	↓	Fair
41	↓	↓	↓	↓	Good
46	Yes	2000	Yes	Yes	Fair
45	↓	↓	↓	↓	Poor
47	↓	↓	↓	↓	Good
48	↓	↓	↓	↓	Excellent
49	↓	↓	↓	↓	Excellent
50	Yes	2200	No	No	Good
51A	↓	↓	↓	↓	Good
52	↓	↓	↓	↓	Excellent
53	↓	↓	↓	↓	Good
54	↓	↓	↓	↓	Fair

[a]At 5500 psi for 2 hr.
[b]2500° C for 2 hr.

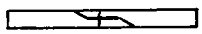
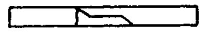
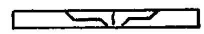
[redacted]

satisfactory results were obtained and most of the joints were rated good to excellent. No preference in bond parameters could be determined from these results. It was found, however, that the insert-type joints were equivalent or superior to the joints containing a single bond. The greatest amount of grain growth across the bond line was observed in the horizontal portion of the joints, perpendicular to the applied force.

Photomicrographs of the cross sections from several of the small butt joints are shown in figure 15; a schematic sketch is presented (fig. 15) to illustrate the location represented by the photomicrograph. The quality of these joints ranged from poor to excellent. An example of a nonbonded region resulting from insufficient plastic flow is shown in figure 15(a). Evidence of a slight edge crumbling problem is shown in figures 15(b) and (c). The bond line of the joint shown in figure 15(d) is very hard to detect, and the joint was classed as excellent.

Bend tests were conducted on the bonded samples in an attempt to evaluate the bond integrity. The small butt joints were cut transverse to the joint into 1/4-inch-wide specimens. The center piece and one end piece were used for testing, and the other end piece was used for metallography. The specimens were tested at either 540° or 650° C.

TABLE VII. - BEND-TEST RESULTS ON VACUUM HOT-PRESS BONDED JOINTS IN TUNGSTEN - URANIUM DIOXIDE COMPOSITES

Specimen	Specimen location	Bend temperature, $^{\circ}C$	Angle of bend, deg	Fracture location
40	Center	540	30	
43	Center		0	
[a]47	Center		20	
[b]54	End		40	
39	End	650	30	
39	Center		25	
41	End		60	
[a]48	End		30	
[b]48	Center		15	
[a]53	End		10	
[b]53	Center		20	
Composite base material	------	540 to 650	90	---------------

[a]Root (bottom of joint) in tension.
[b]Face (top of joint) in tension.

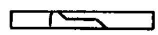
The results of the bend tests, including sketches illustrating the location of failures, are presented in table VII. All the samples failed during the bend test at both temperatures, but metallographic examination of the fractures indicated that in all cases the fracture did not follow the bond line. Although no real evaluation of bond quality could be determined, these results indicate that the bonding time-temperature cycle and/or a contamination problem (which will be discussed in the section Contamination problems) resulted in an increase in the transition temperature of the W-UO_2 base material because the untreated base material can be easily bent at temperatures as low as 400° C. The edge crumbling condition observed in figure 15(b) also could have contributed to the poor bend ductility.

Tensile blanks. - Seven tensile blanks, representative of each of the four bonding and heat-treating conditions, were machined to the tensile test configuration used and were tested at 2500° C. Both the bonding conditions and tensile data are presented in table VIII. The strengths obtained on these joints compare favorably with the strength of the unbonded W-UO_2 base material. The insert-type specimens bonded at 2000° C with a 2500° C postbonding heat treatment had the highest strength, but the strongest, specimen 38, unexpectedly showed evidence of failure along the bond line.

Cylinder butt joints. - Six cylinders were evaluated visually and metallographically and were checked for dimensional uniformity. The bonding parameters and dimensional data for these cylinders are listed in table IX. Only specimen G was essentially circu-

TABLE VIII. - TENSILE-TEST RESULTS FOR VACUUM

HOT-PRESS BONDED TENSILE BLANKS

Specimen	Insert	Bonding temperature,[a] °C	Postbonding thermal treatment[b]		Ultimate tensile strength at 2500° C, psi	Fracture location
			Temperature, °C	Time, hr		
14	No	2200	2500	2	3270	
28	No	2200			3380	
36	Yes	2000			3880	
38		2000			4210	
29		2200			3040	
40		2200	2000		3380	
56		2200	2000		3600	
Composite base material	---	----	----	-	3400 to 4400	---------------

[a]At 5500 psi for 2 hr.

[b]All samples tungsten clad by vapor deposition prior to heat treating.

[redacted]

TABLE IX. - DIMENSIONAL INSPECTION RESULTS FOR VACUUM

HOT-PRESS BONDED CYLINDERS[a]

Specimen	Tungsten coated	Postbonding thermal treatment		Cylinder diameter, in.		Wall thickness, in.	
		Temperature, ^{o}C	Time, hr	Maximum	Minimum	Maximum	Minimum
I	Yes	2000	2	0.567	0.554	0.031	0.028
O	Yes	2000	2	.559	.547	.033	.026
F	No	None	None	.542	.535	.024	.021
G	↓	↓	↓	.541	.540	.020	.016
P	↓	↓	↓	.547	.540	.021	.020
R	↓	↓	↓	.559	.545	.037	.024

[a]All cylinders bonded at 2200^{o} C and 5500 psi for 2 hr.

lar; all the other cylinders were somewhat out of round. Because the out-of-roundness appeared to be a characteristic of the process, an attempt was made to determine the maximum variation in diameter. Also, only specimen P had a nearly constant wall thickness. These dimensional variations are apparently the result of the deformation and plastic flow which occured during bonding. Variation in wall thickness also resulted from postbond tungsten cladding by vapor deposition and sticking of the tantalum carbide with subsequent nonuniform removal techniques.

Cross sections were taken from each cylinder for metallographic examination at locations which were suspect on the basis of visual and radiographic examination. All the joints were rated poor except for the joint in specimen O which was rated good (fig. 16). Except for one small unbonded area in the section examined, extensive grain growth was achieved across the interface. Unfortunately, major defects were detected visually at two locations away from the joint as a result of an arcing condition (ref. 4) that occurred during bonding, and holes were burned through the composite.

The most common flaw detected in the other specimens was the lack of grain growth across the interface. A typical flaw of this type is shown in the photomicrograph in figure 17(a). Other types of more severe joint defects are shown in figure 17(b) and (c). The joint shown in figure 17(b) is not intact which has resulted in an extensive loss of UO_2. Excessive plastic flow occurred in the specimen shown in figure 17(c), and gross defects resulted.

T-joints. - Vacuum hot-press bonding of T-joints was not very successful. The three T-joints received for evaluation were bonded at 2000^{o} C and 5500 psi for 1 hour. No postbonding heat treatment was employed. Examination of these joints indicated that there was a tendency for bending of the horizontal members during bonding. The major deficiency of all the joints was that bonding was achieved over a small percentage of the

[redacted]

TABLE X. - CARBON CONTENT OF VACUUM HOT-PRESS BONDED TUNGSTEN-URANIUM DIOXIDE SPECIMENS[a]

Cylinders[b]		Small butt joints[c]	
Specimen	Carbon content, ppm	Specimen	Carbon content, ppm
O	1590	41	20
I	97	41	59
P	570	43	50
F	240	43	58
G	226	49	100
G	61	49	79
R	>2500	49	26
		52	390

[a]Unbonded base material carbon content, 10 ppm.
[b]Samples used for analysis were 1/8-in. -wide rings cut from central portion of cylinders.
[c]Samples were 1/4- by 1-in. slices taken at midlengths of joints except for specimen 43 which taken from one end.

bond interface. A typical joint, illustrating this type of defect, is shown in figure 18.

Contamination problems. - Contamination of the W-UO_2 composites was a major problem during vacuum hot pressing because of the close proximity of the carbon dies. To minimize the carbon contamination problems, the graphite dies were separated from the W-UO_2 composites by a layer of tantalum carbide. Unfortunately, this carbide tends to stick to the composites (as shown in figs. 16 and 17(c)), and thus the bonded joints can be contaminated with tantalum carbide.

A grinding operation is often necessary in order to remove the carbide from the composite. It is quite difficult to remove all the carbide from the flat plates without damaging the tungsten cladding, and it is virtually impossible to grind all the carbide from bonded cylinders that are not round.

Chemical analysis of both bonded cylinders and plates verified that the more severe carbon contamination occurred in the cylinders. These data are presented in table X. Two of the cylinders (O and R) contained over 1500 parts per million carbon. The photomicrograph of cylinder O (fig. 16) shows that all the tantalum carbide was not removed from the inside diameter of the cylinder prior to the vapor deposition of a tungsten coating. A similar tantalum carbide layer was observed on cylinder R (fig. 17(c)). Examination of cylinder P by X-ray fluorescene techniques indicated that tantalum (believed to be in the form of tantalum carbide) was present in the vicinity of the joint and at 180° from the joint. These two areas were the only areas examined.

Discussion. - The feasibility of using the vacuum hot-press bonding technique to weld W-UO_2 composites was established. Unfortunately, the generally satisfactory results obtained on the small butt joints and tensile blanks were not reproduced in the cylinder and T-joint configurations, where only partial bonding was achieved.

No preference in bonding condition could be determined metallographically between specimens bonded at 2000° C with a 2500° C postbonding heat treatment and specimens bonded at 2200° C without a postbonding heat treatment. Tensile tests at 2500° C, how-

[redacted]

ever, indicated that the former bonding procedure might yield the stronger joints. Both the single butt-joint design and the more complex insert-type joint yielded similar results. The bend tests conducted at 540° and 650° C were not too meaningful in distinguishing between different bonding parameters because all the samples failed in a brittle manner. The fractures, however, did not follow the bond line in any of the bend tests.

Contamination of the $W-UO_2$ composites by tantalum carbide was a major problem, and methods were not developed to ensure complete removal of the tantalum carbide layer. Therefore, further development of this welding process is required before it could be considered for fuel element joining applications.

Vapor Deposition Welding

Small butt joints. - Five vapor deposition welded, small butt joints were examined by using both the dye-penetrant and the radiographic methods. These were the only specimens that were joined by this process and submitted for evaluation. Generally, there was good correlation between nondestructive test methods, and four of the five joints were found to contain centerline defects. This type of defect was the most recurrent type of defect encountered in the program. A typical defect of this type is shown in figure 19.

Another type of defect, characteristic of the results of this vapor deposition process, is shown in figure 20. At midlength of the joint (fig. 20(a)) the grain size of the deposited W was quite small, and the joint was defective at this location. At another location in the same joint (fig. 20(b)), the grain size was considerably larger and there were no visible defects. At both locations, excellent bonding occurred between the vapor deposited tungsten and the composite base material, and grain growth occurred across the interface. This joint was heat treated at 1925° C in vacuum after welding (as were all the five samples that were submitted), and this heat treatment apparently was effective in promoting grain growth across the centerline of the joint, as shown in figure 20(b).

The only specimen that was free of centerline defects exhibited an offset condition more severe than that shown in figure 20(b). This sample also exhibited a significant variation in grain size along the joint.

Discussion. - Evaluation of the five small butt joints indicated that the feasibility of vapor deposition welding of the composite was not established during this program. Four of the five joints contained cracks or voids; the fifth sample was poorly alined and had variable grain size. The factors which control the occurence of centerline defects and variable grain size were not characterized. Thus, on the basis of the unpromising results obtained, work on this process was terminated at the conclusion of the initial

[redacted]

feasibility study. Although vapor deposition welding has potential for joining $W\text{-}UO_2$ composites, considerably more development of the process is required.

Gas Tungsten-Arc Brazing

Small butt joints. - Nineteen small butt joints brazed with a gas tungsten-arc heat source were evaluated. The results of dye-penetrant examination of these joints are given in table XI. Two of the four joints brazed with the tungsten - 50-percent molybdenum - 3-percent rhenium (W - 50 Mo - 3 Re) alloy and nine of the fifteen joints brazed with the tungsten - 25-percent osmium (W - 25 Os) alloy were cracked. Several of the defects were confirmed metallographically, as indicated in table XI.

Typical photomicrographs of a good (except for misalinement) and of a cracked small butt joint brazed with the W-50 Mo-3 Re alloy are shown in figure 21. The loss of UO_2 which was observed in all the brazed joints is probably a result of the severe thermal cycle to which the composites were exposed during gas tungsten-arc heating. The inter-

TABLE XI. - DYE-PENETRANT EXAMINATION RESULTS OF SMALL BUTT JOINTS BRAZED WITH GAS TUNGSTEN-ARC PROCESS[a]

Specimen	Braze alloy	Defect indications
276	Tungsten - 50-percent molybdenum - 3-percent rhenium	None
282		None
288		Centerline crack[b]
289		Cracks[b]
295	Tungsten - 25-percent osmium	Cracks[b]
296		None
297		Cracks
298		None
299		Fine cracks[b]
301		Fine cracks
303		None
304		Cracks[b]
305		None
306		None
308		None
309		Fine cracks[b]
310		Fine cracks
314		Fine cracks[b]
355		Fine cracks

[a]Start defects, crater defects, and small pits were ignored.
[b]Confirmed by metallography.

[redacted]

granular nature of a typical defect (fig. 21(b)) can be easily seen. Intergranular cracking of this type was a persistant problem for both the braze alloys containing molybdenum; that is, W-50 Mo-3 Re and Mo-5 Os. The reason for this difficulty is believed to be associated with the fact that molybdenum has an extremely low solubility for oxygen (about 1 ppm), and thus oxide phases are always present (ref. 8). The oxygen reacts to form a molybdenum - molybdenum oxide eutectic which segregates at the grain boundaries. It is thought that this eutectic was in the form of a thin film along the grain boundaries. The lower melting eutectic remained molten for a sufficient period of time after the braze metal had solidified to produce a situation wherein shrinkage stresses were applied normal to the molten grain boundaries with the resulting intergranular cracking.

A different type of cracking was observed in the small butt joints brazed with the W-25 Os. A longitudinal section through the centerline of this brazed joint is shown at two magnifications in figure 22. Because of the semiregular pattern of the cracking, and because it is apparently not intergranular in nature, it appears that "cold" cracking occurred far below the solidus temperature due to stresses that developed upon cooling to room temperature. The high magnification photomicrograph (fig. 22(b)) shows that the progression of the crack is almost totally within the region which is believed to be primarly the σ-phase (ref. 9). The cracks tended to avoid the primary crystals of the tungsten rich α-phase, and it would appear that the α-phase was comparatively ductile. The cracking problem in the W - 25 Os alloy might be reduced by using an alloy richer in tungsten than the W - 25 Os composition.

Bend tests were conducted on 1/4- by 1-inch pieces cut transverse to the joints in two W - 25 Os samples and in one W - 50 Mo - 3 Re sample. Excess braze metal in the joint region was not removed prior to bending for fear that cracking might be induced. The braze metal resisted deformation which resulted in sharp bend angles in the composite base material immediately adjacent to the braze metal. Although no qualitative data could be obtained on braze metal ductility, these results indicate that the base material is not embrittled by the brazing operation.

Tensile blanks. - No defects were detected by dye-penetrant examination of 12 tensile blanks brazed with the W - 25 Os alloy. Photographs of the face and root of a typical brazed specimen are shown in figure 23. Some excess braze metal is evident at the joint. This excess metal was removed by grinding to within 0.001 inch of the major surfaces of the plates before the specimens were machined to the tensile configuration.

Duplicate tensile tests were conducted on as-brazed samples at 2500° C and the results of these tests are presented in table XII. Fracture occurred in the composite plates at a stress level close to the average strength of the unbrazed composite plate; that is, 3970 psi at 2500° C. Because the braze metal was somewhat greater in cross-sectional area, the stress at fracture was 15 to 20 percent less than that in the base material. Thus, it cannot be stated that the braze metal is stronger than the composite

[redacted]

TABLE XII. - TENSILE PROPERTIES OF TUNGSTEN - 25-PERCENT

OSMIUM BRAZED TUNGSTEN - URANIUM DIOXIDE

COMPOSITES TESTED AT 2500° C

Specimen	Condition	Ultimate tensile strength, psi	Fracture location
334	As-brazed	3630	Base material, 1/4-in. from braze
343	As-brazed	3370	Base material, 1/8-in. from braze
Composite base material	Unbrazed	3400 to 4400	--------------------

even though fracture occurred in the base material.

In order to simulate the intended service application, a brazed joint was heated for 10 hours at 2500° C in flowing hydrogen after which it was examined metallographically. Duplicate cross sections were taken 3/8 inch apart (3/16-in. on either side of midlength of the joint). The resulting microstructure (fig. 24(a)) was generally improved in that the grains of the composite base material and braze metal grew together into a single (α-phase) structure within the joint. The same diffusion effect may be observed in another part of the joint by noting in figure 24(a) that the interface between the tungsten-rich α-phase and the σ-phase is moving upward at the face (top) of the joint; that is, the σ-phase is transforming to the α-phase as the osmium content is reduced by diffusion. At the root (bottom) of the joint shown in figure 24(a) only a thin stringer of σ-phase remains in the α-matrix. The reason for the presence of braze metal porosity shown in the upper part of figure 24(a) is unknown. In the cross section shown in figure 24(b), the same diffusion effects can be seen; that is, the σ-phase matrix of the braze metal is in the process of being converted to tungsten-rich α-phase. This reaction is incomplete within the joint because the gap between the mating surfaces was apparently greater at this location (fig. 24(b)) than it was in the section shown in figure 24(a). No evidence of Kirkendall-type porosity was observed, indicating that the diffusion rates of osmium and tungsten across the composite-braze metal interface are similar.

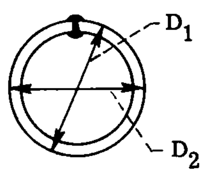
Room temperature microhardness determinations were helpful in microstructural analysis of the braze metal shown in figure 24. Diamond pyramid hardness (DPH) of the tungsten matrix of the composite base material was quite low (350 DPH) compared with σ-phase regions in the braze metal (calculated values as high as 1600 DPH). Hardness of α-phase regions in the braze metal generally was on the order of 800 DPH although at midthickness of the cross section shown in figure 24(a) the hardness was 440 DPH presumably because of comparatively low osmium content.

TABLE XIII. - INSPECTION RESULTS OF BRAZED

TUNGSTEN - URANIUM DIOXIDE CYLINDERS

Specimen	Braze alloy	Dimensions,[a] in.		Defect indication[b]
		D_1	D_2	
369C	Molybdenum - 5-percent osmium	0.542	0.535	Transverse crack
370C	Molybdenum - 5-percent osmium	.538	.534	None
372C	Tungsten - 50-percent molybdenum - 3-percent rhenium	.538	.531	↓
358C	Tungsten - 25-percent osmium	.540	.532	↓
359C	↓	.540	.536	↓
360C	↓	.539	.532	↓
361C	↓	.538	.534	↓
362C	↓	.537	.534	↓
364C	↓	.540	.531	↓
365C	↓	.542	.537	↓

[a]Wall thickness, 0.020 to 0.021 in.; length, 3/4-in.



[b]Obtained by dye-penetrant examination. Start defects, crater defects, and small pits were ignored.

Cylinder butt joints. - Dye-penetrant examination of the cylinder butt joints indicated that 9 of the 10 joints evaluated were defect-free, as shown in table XIII. The defective joint was in one of the two cylinders brazed with the Mo - 5 Os alloy. Included in table XIII are the results of diametral measurements, which indicate that the majority of cylinders were somewhat out-of-round. At the current stage of development, however, this degree of distortion is not considered excessive.

Cylinders brazed with each of the three alloys are shown in figure 25. In this figure it can be clearly seen that the amount of braze metal at the joints was excessive and the braze surfaces were irregular and rippled. The uneven braze distribution was a result of the method of placing the braze alloy in the joint by using fragments of the alloy. The use of braze alloys in the form of wire or foil should result in a more uniform braze distribution.

A typical cross section of a W-UO_2 cylinder brazed with the W - 25 Os alloy is shown in figure 26. The braze alloy in this joint exhibited excellent flow and wetting action, and no cracks or porosity were evident. The microstructure differs markedly from that shown in figure 24 because the structure in figure 26 is as-brazed. A 10-hour heat treatment at 2500° C would result in a microstructure similar to that shown in figure 24. An excessive amount of braze metal was present at the joint shown in figure 26 and some dissolution of the composite occurred. In addition, the joint was not perfectly alined.

A nearly ideal W - 25 Os braze joint is shown in figure 27 in the as-brazed condition. The joint gap was relatively small and only a minimum amount of excess braze metal was present. This joint was not typical of those produced in this program, but it indicates what might be obtained with additional process development. It is evident in figure 27 that the edges of the composite were in the warm sheared condition.

T-joints. - T-joints in W-UO_2 composites were not successfully fabricated by brazing with a gas tungsten-arc heat source. Joint accessability was a major problem, and all attempts to join the members resulted in burn-through of the W-UO_2 composites.

Discussion. - The program to develop suitable alloys for use with a gas tungsten-arc heat source in order to braze W-UO_2 composite showed feasibility despite the relatively crude manner in which the braze alloy was applied. The two-phase W - 25 Os braze alloy showed the most promise of the alloys investigated. The cold cracking problems encountered with this alloy could possibly be eliminated, or at least minimized by using holding fixtures that would reduce the restraint and quenching during brazing. In addition, preheating and/or postbrazing heat treatments and a more uniform distribution of braze metal at the joint also might reduce the cold cracking.

A considerable amount of intergranular cracking was encountered with the two most promising single-phase braze alloys, W - 50 Mo - 3 Re and Mo - 5 Os. This type of cracking is believed to be due to the presence of a molybdenum - molybdenum oxide eutectic at the grain boundaries. A possible source of oxygen contamination could have been the UO_2 in the composites, if the brazing produced dissolution of the composites.

The results of high-temperature tensile tests on W-UO_2 composites, brazed with the W - 25 Os alloy, indicated that in the as-brazed condition the joints were strong in that failure occurred away from the joint in the base material. A postbrazing heat treatment generally improved the joint region by promoting grain growth across the interfaces by a diffusion mechanism in which the σ-phase matrix of the braze metal was transformed to the more crack resistant α-phase. No Kirkendall-type porosity was observed.

Special Electron Beam Welding Techniques

Small butt joints. - Several butt joints were produced by using a special electron beam welding technique. In this work it was found that alinement of the electron beam was a major problem because the slightest deviation from the joint centerline would result in the beam impinging on the W-UO_2 composite which, in turn, would result in defects in the joint.

Three cross sections from one of the more promising joints are shown in figure 28. The electron beam was off-center, particularly near the start. From the limited amount of work that was done, it appears that an unfueled zone of about 0.030 inch would be required so that the electron beam would remain within the plasma sprayed tungsten. Some cracking (fig. 28(b)) and separation of the cladding and core (fig. 28(a) and (c)) can also be seen. A metallurgical bond was achieved in the sample between the plasma sprayed tungsten and the composite as evidenced by the substantial grain growth across the interface. Porosity is present in the plasma sprayed tungsten; however, this is a characteristic of this material.

Discussion. - The feasibility of joining W-UO_2 composites by using the special electron beam technique was not demonstrated in this study since no defect-free joints were produced. The results indicated that an excessively wide unfueled zone would be required to consistently produce high quality joints. For these reasons and because other welding techniques showed more promise, work on this program was terminated after the initial studies.

COMPARISON OF WELDING PROCESSES

Several of the welding processes studied in this investigation were successful to varying degrees in joining W - 20-volume-percent UO_2 composites and in meeting the goals of the program. The most success was attained with the gas-pressure bonding process. Well bonded, undistorted joints were produced in all the required configurations with this process. A slight alinement problem was evident in the cylinder joints, but it is believed that this problem could be solved with further work. A high degree of joint integrity was demonstrated on the small butt joints by successful bend tests at 650^{o} C. The results of tensile tests at 2500^{o} C showed that the strength of the gas-pressure bonded joints is equivalent to that of the unbonded W-UO_2 plate, although in all cases fracture occurred along the bond line with very little deformation. The joints also were very promising in that no unfueled region was produced at the joint and clad-to-clad bonding was achieved. The major drawback to the gas-pressure bonding technique is the need for numerous time-consuming operations and associated costs necessary to produce the joints.

The W-UO_2 composites also were successfully joined by the gas tungsten-arc brazing process by using a W - 25-weight-percent Os braze alloy. Sound joints were readily produced in the cylindrical butt-joint and tensile-blank configurations. More difficulty was encountered in producing sound joints in the small butt configuration because of fixturing restraint. An accessibility problem prevented brazing of the T-joint configuration. Because of the relatively crude method employed to preplace the braze alloy, there was generally an excessive amount of braze metal present in the immediate vicinity of the joints. It is thought, however, that this problem could be minimized with more refined techniques. Unfueled zones 0.005-inch or less in width were readily obtained with this welding process. High-temperature (2500^{o} C) tensile test results indicated that the brazed joints had good strength because the tensile fractures occurred in the W-UO_2 base metal. High-temperature treatment of the joints at 2500^{o} C in flowing hydrogen for 10 hours tended to improve the joint region by promoting diffusion of the braze alloy and grain growth across the braze metal and base material interfaces.

The magnetic force welding process showed considerable promise because the welding operation takes place in a fraction of a second with no loss of fuel at the joint and no unfueled region is produced. However, there is metal upset at the joint which must be controlled within acceptable limits. Butt joints in flat plates and in cylinders were produced, but the persistence of a cracking problem in the composites, transverse to the joints, negated the positive features. If the cracking problem could be overcome and the metal upset satisfactorily controlled, this joining method would be very promising because of the high production rates that could be achieved.

The feasibility of joining W-UO_2 composites by using a vacuum hot press was established for bonding parameters of 2000^{o} to 2200^{o} C and 5500 psi for 2 hours. Unfortunately, the generally satisfactory results which were obtained for the small butt joints and the tensile blanks were not reproduced in the cylindrical or T-joint configuration. At the heart of this problem is the difficulty involved in attempting to control completely the small amount of plastic deformation that is necessary in this process in order to achieve complete welding at the mating surfaces. In addition, severe contamination from the fixturing also was encountered. This process does not appear to be as promising as the three previously discussed processes because it poses several difficult problems and does not seem to offer any advantages over the other processes.

Very little success was attained with either vapor deposition welding on the special electron beam welding techniques. Neither process produced defect-free joints in the composite plates, and both programs were terminated after the initial feasibility study. Another drawback to the use of these processes is the presence of a relatively wide unfueled zone. If the problems associated with vapor deposition welding can be overcome, this method does offer promise because welding can be achieved at relatively low temperatures.

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CONCLUSIONS

The results of this investigation have shown that only one of the six welding processes studied met all of the program goals in joining tungsten - 20-volume-percent uranium dioxide composites. This solid-state welding process, gas-pressure bonding, was used to produce usable joints in all the required configurations with complete bonding at the mating surfaces, no unfueled zone, and elevated temperature strength equivalent to that of the base material.

Other processes which were less successful but demonstrated a great deal of promise are the gas tungsten-arc brazing technique that uses the tungsten - 25-weight-percent osmium alloy and the magnetic force welding process. Further development of these welding techniques appears warranted.

A vacuum hot-press bonding technique was generally successful only with joints in the flat-plate configuration. The results obtained on cylinder seam butt joints and T-joints were unsatisfactory.

The vapor deposition welding and special electron beam welding techniques studied did not produce defect-free small butt joints. It is possible that, with further efforts, sound joints could be produced, but the relatively wide unfueled zone would continue to be a major drawback to these welding techniques.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, February 23, 1967,
122-28-01-01-22.

REFERENCES

1. Watson, Gordon K.: Fabrication of Thin Tungsten-Uranium Dioxide Composite Plates. NASA TM X-1073, 1965.

2. Peterson, J. H.; Meiners, K. E.; and Gripshover, P. J.: Joining of Tungsten-UO_2 Composites by Diffusion Bonding. Battelle Memorial Institute (NASA CR-54791), Oct. 15, 1965.

3. Shaw, C. H.; and Boolen, R. F.: Magnetic Force Welding of Tungsten-Uranium Dioxide Composites. Rep. No. BNWL-286 (NASA CR-54839), Battelle-Northwest, Jan. 24, 1966.

[redacted]

4. Weinberg, A. F.; and Lindgren, J. R.: Joining of Tungsten-Uranium Dioxide Composite by Diffusion Bonding. Rep. No. GA-6602 (NASA CR-54842), General Atomic Div., General Dynamics Corp., Aug. 12, 1965.

5. Japka, J. E.; and Gordon, E.: Joining of Tungsten-Uranium Dioxide Composites by Gas Tungsten-Arc Brazing. United Nuclear Corp.-Development Division (NASA CR-54841), May 1966.

6. Grisaffe, S. J.; and Spitzig, W. A.: Observations on Metallurgical Bonding Between Plasma Sprayed Tungsten and Hot Tungsten Substrates. ASM Trans., vol. 56, no. 3, Sept. 1963, pp. 618-628.

7. Buzzard, Robert J.; and Gill, Frank L.: High-Temperature Mechanical Properties of a Tungsten-Uranium Dioxide Composite. NASA TM X-1018, 1964.

8. Thompson, E. G.: Welding of Reactive and Refractory Metals. Bull. No. 85, Welding Research Council, Feb. 1963.

9. English, J. J.: Binary and Ternary Phase Diagrams of Columbium, Molybdenum, Tantalum, and Tungsten. Rep. No. 152, Defense Metals Information Center, Apr. 28, 1961.

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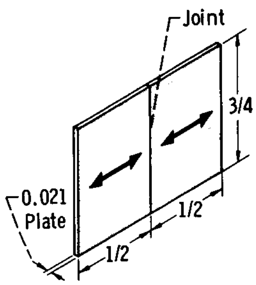
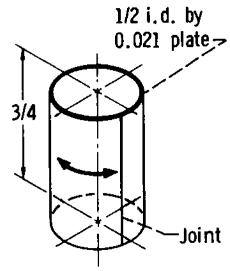
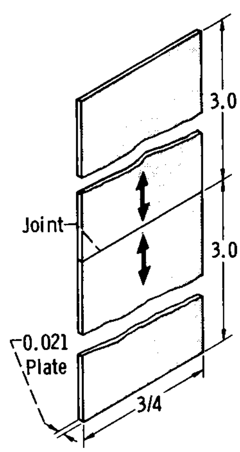


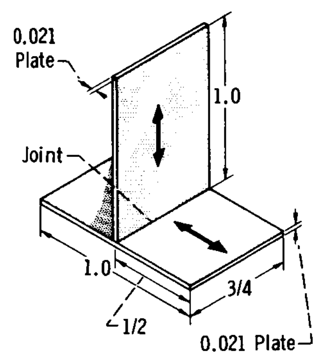
(a) Small butt joint. Quantity required: phase I, 5; phase II, 18.



(b) Butt joint in cylinders. Quantity required: phase II, 6.



(c) Tensile blanks. Quantity required: phase II, 12.



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(d) T-joint. Quantity required: phase II, 6.

Figure 1. - Required joint configurations. (Bold arrows indicate rolling direction for tungsten - uranium dioxide composite plate. All dimensions are in inches.)

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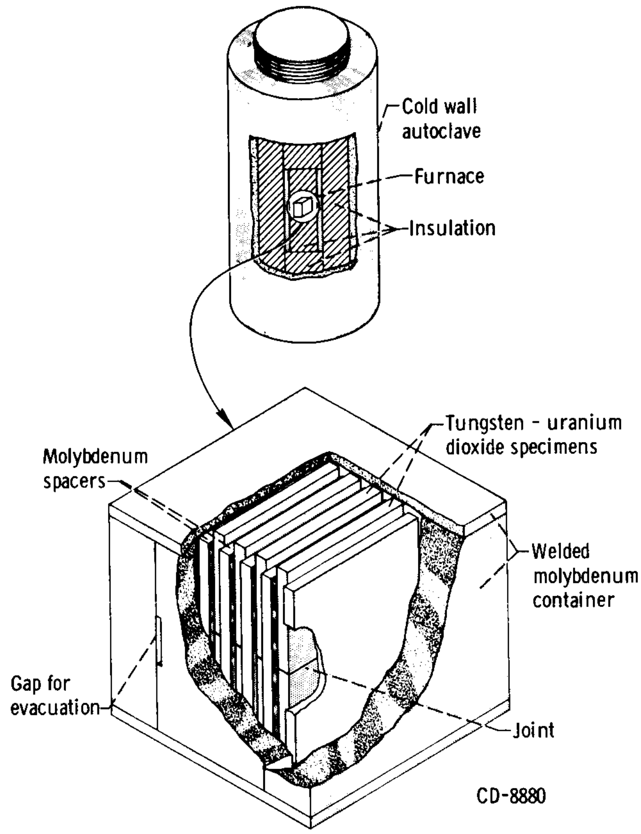

Figure 2. - Gas-pressure bonding autoclave and container for small butt joints.

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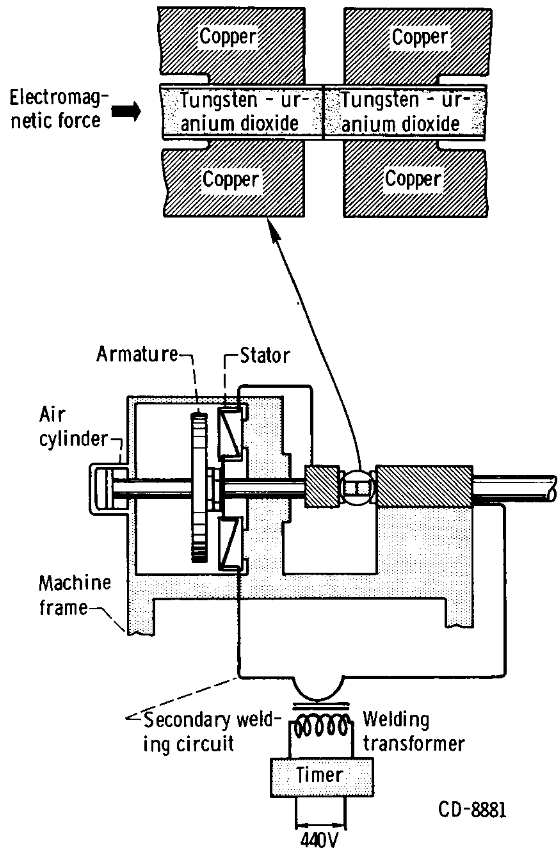

Figure 3. - Magnetic force welding machine and specimen fixturing arrangement for small butt joints.

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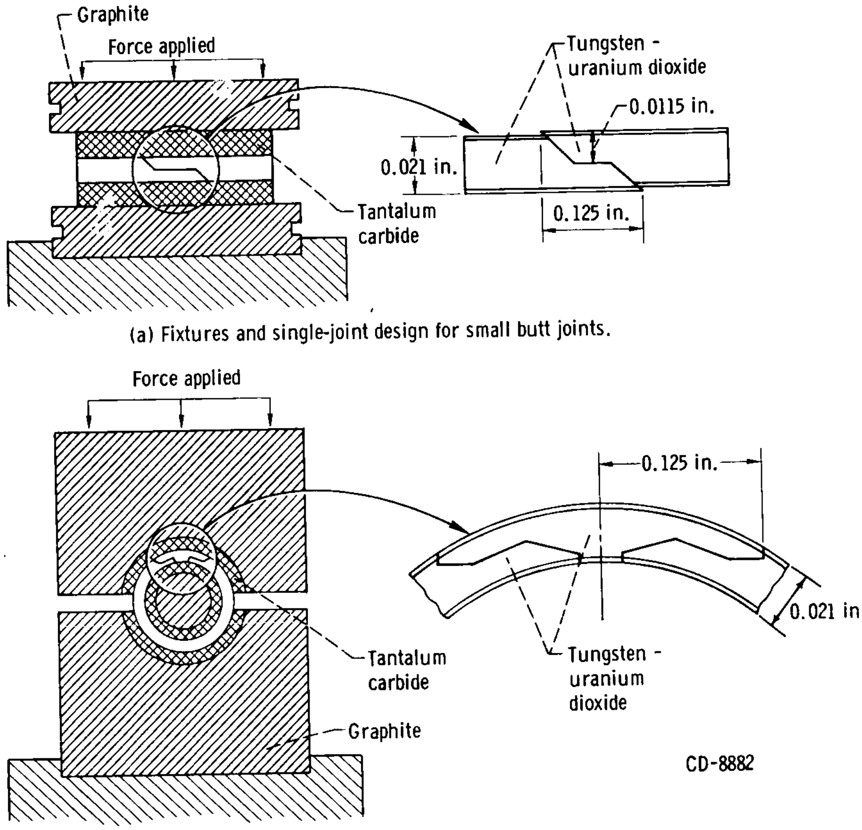

(a) Fixtures and single-joint design for small butt joints.

(b) Fixtures and insert-type joint design for cylinders.

Figure 4. - Bonding fixtures and joint designs for vacuum hot-press bonding.

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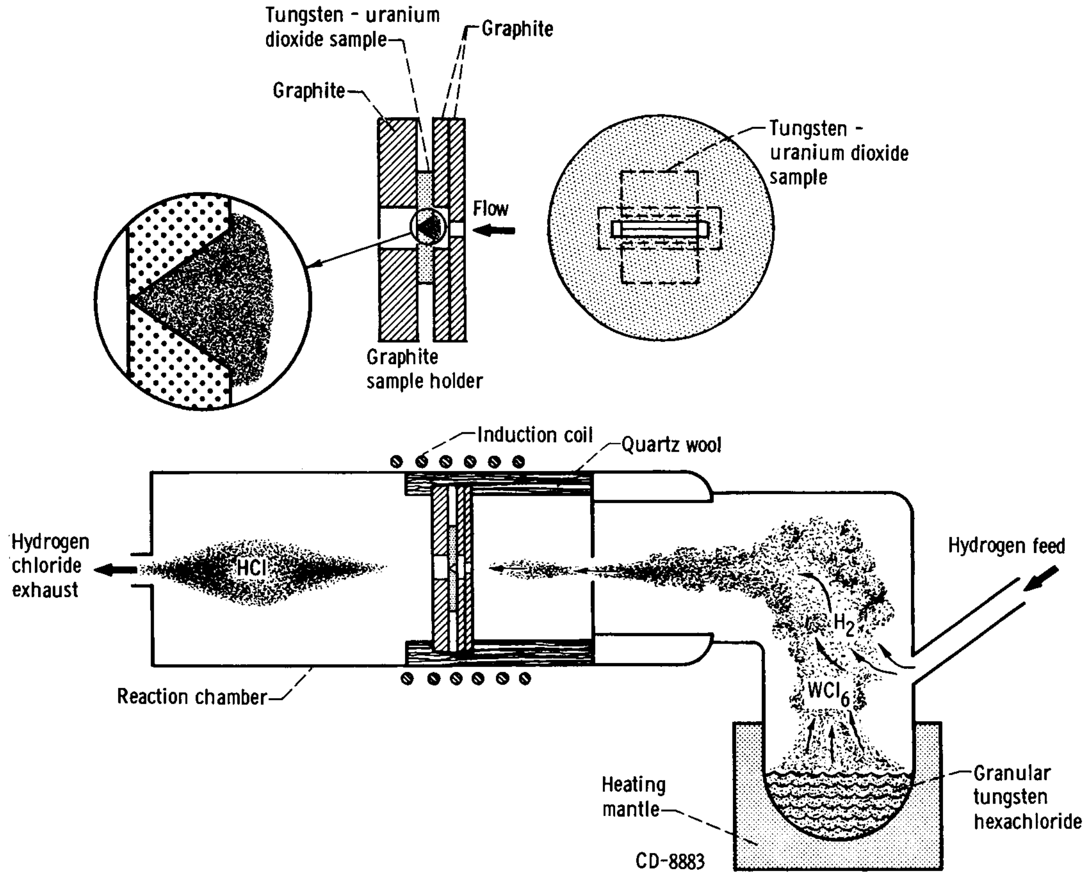

Figure 5. - Apparatus and sample holders for vapor deposition welding.

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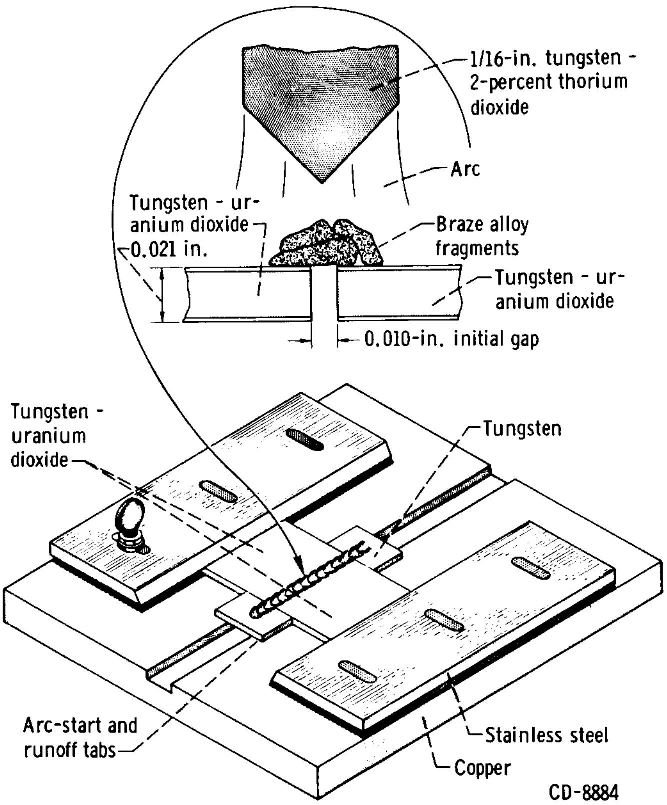

Figure 6. - Sample holder arrangement for gas tungsten-arc brazing small butt joints.

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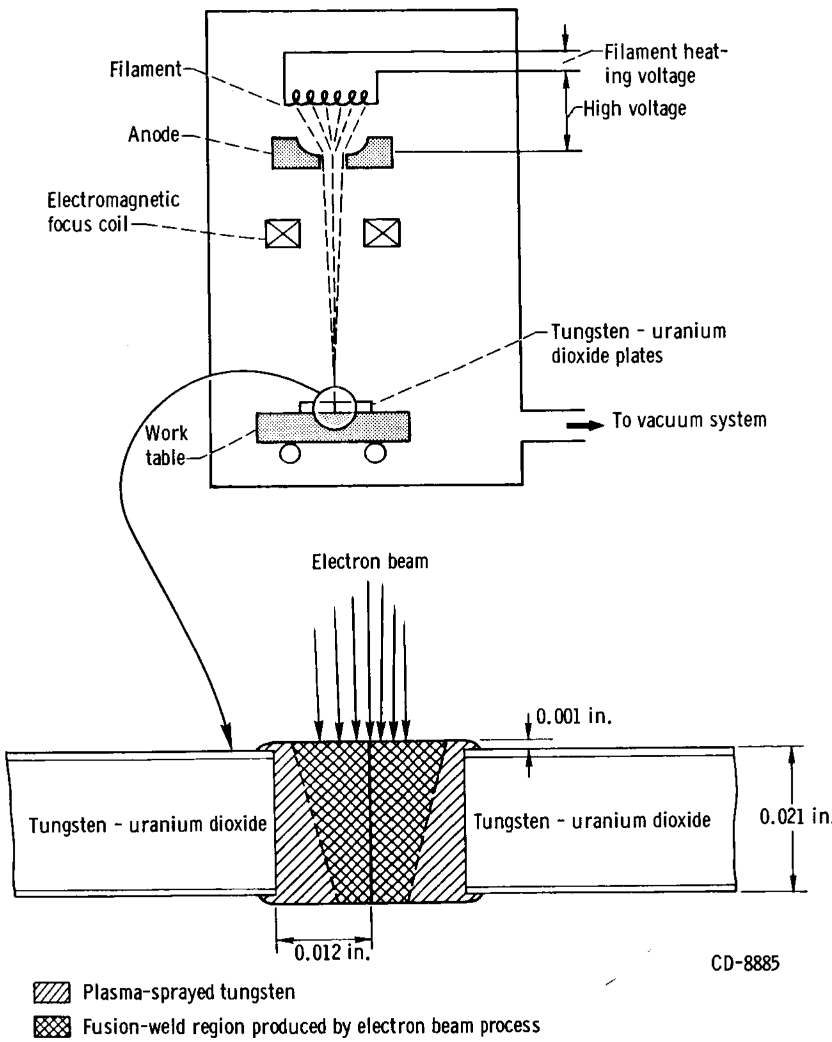

Figure 7. - Schematic illustration of edge cladding and subsequent electron beam fusion welding technique.

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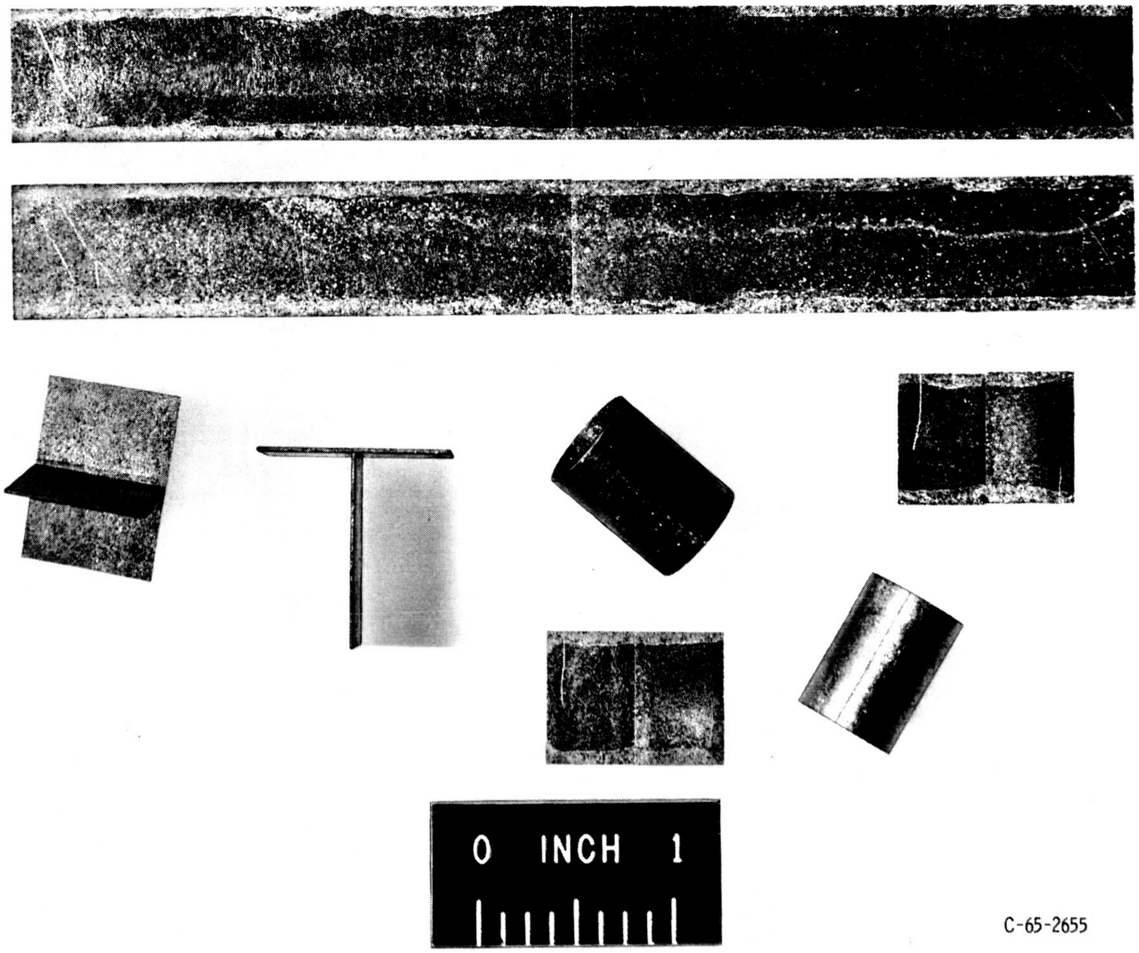

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Figure 8. - Typical gas-pressure bonded specimens of four configurations required.

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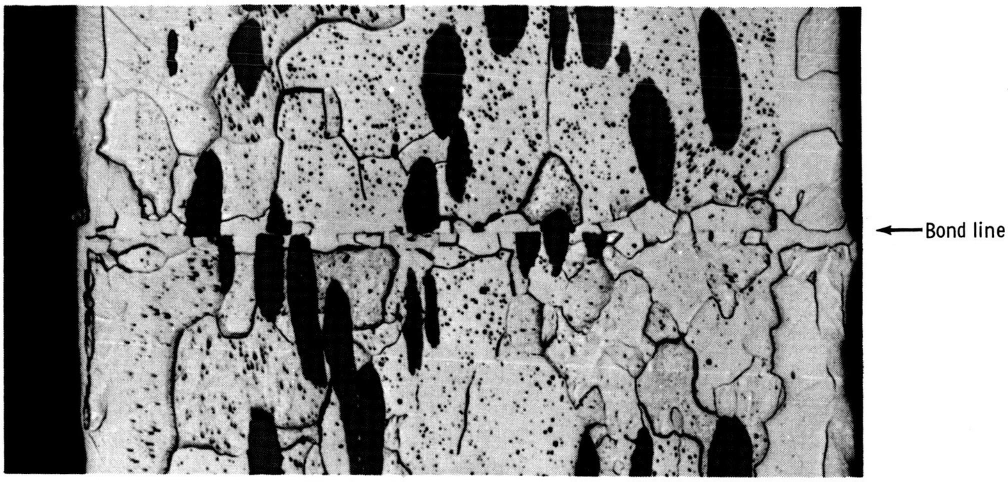
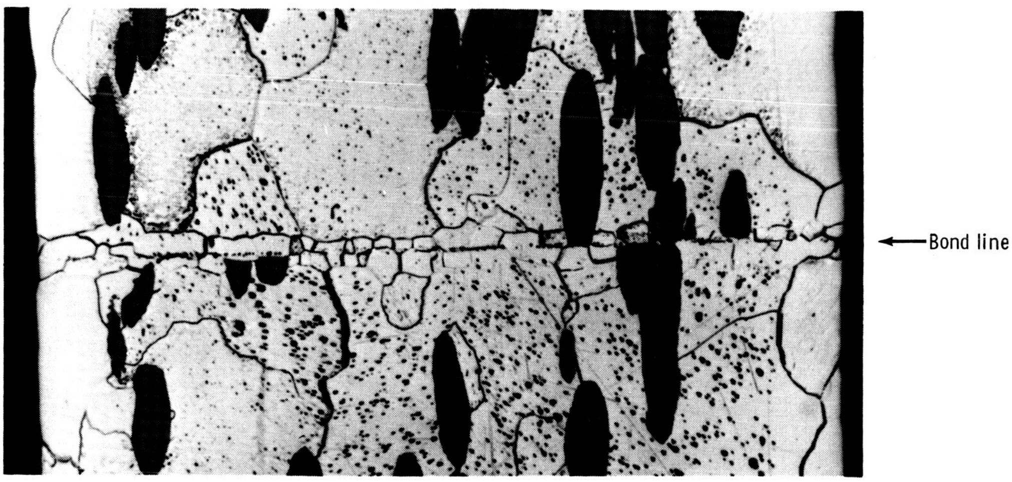
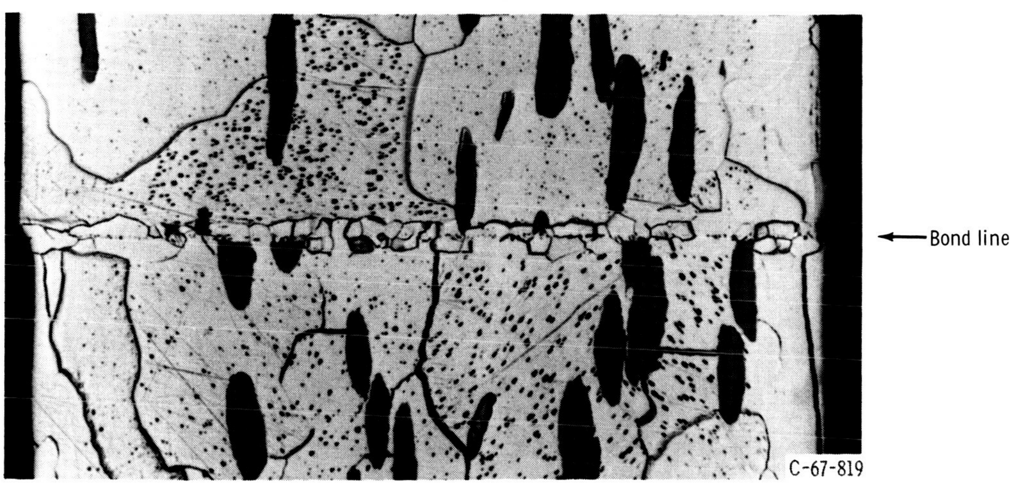


(a) No powder at interface. Sample 6 in as-bonded condition.



(b) Powder applied. Sample 10 in as-bonded condition.



(c) Powder applied. Sample 18 heat treated at 2040° C for 2 hours in vacuum.

Figure 9. - Cross sections of gas-pressure bonded small butt joints. Etchant, Murakami's reagent. Magnification, X200.

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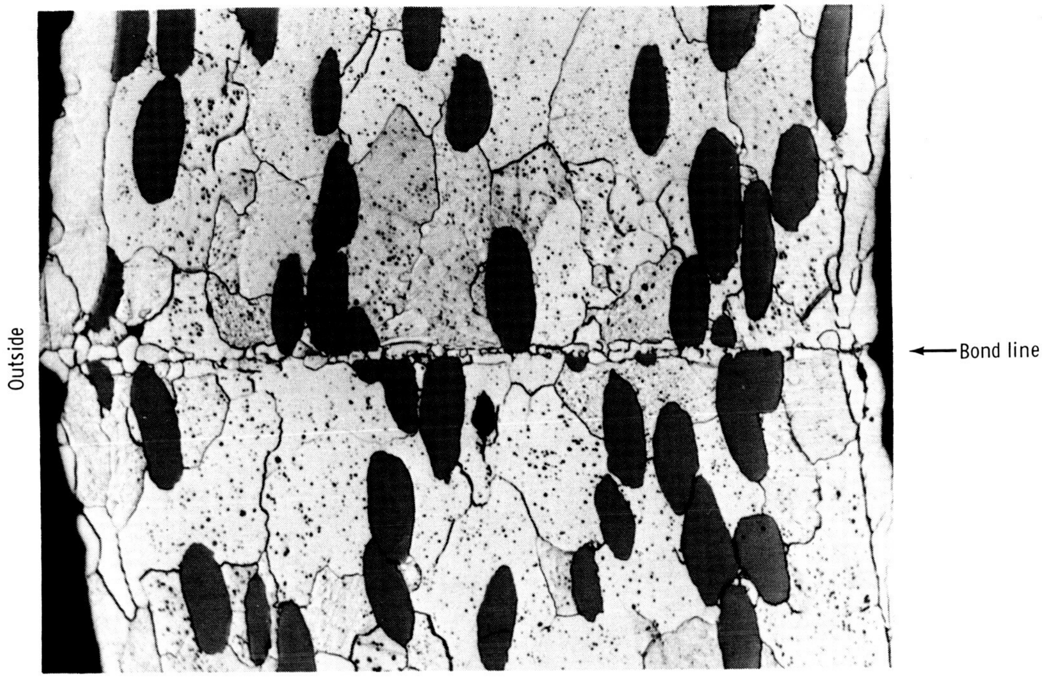
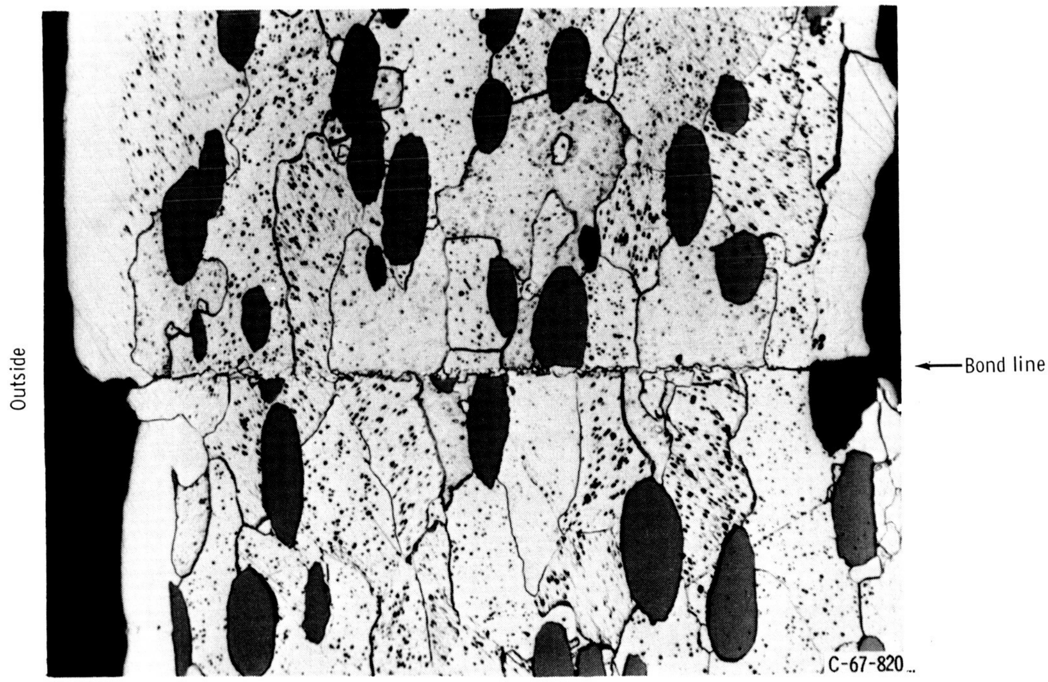


(a) Powder applied. Cylinder 9, in as-bonded condition bonded as full cylinder. (Fine grains present along bond line. Alinement, excellent.)



(b) Powder applied. Cylinder 4, in as-bonded condition bonded in halves. (Defect evident at right side of joint, i.e., inner surface.)

Figure 10. - Cross sections of gas-pressure bonded cylinders. Etchant, Murakami's reagent. Magnification, X200.

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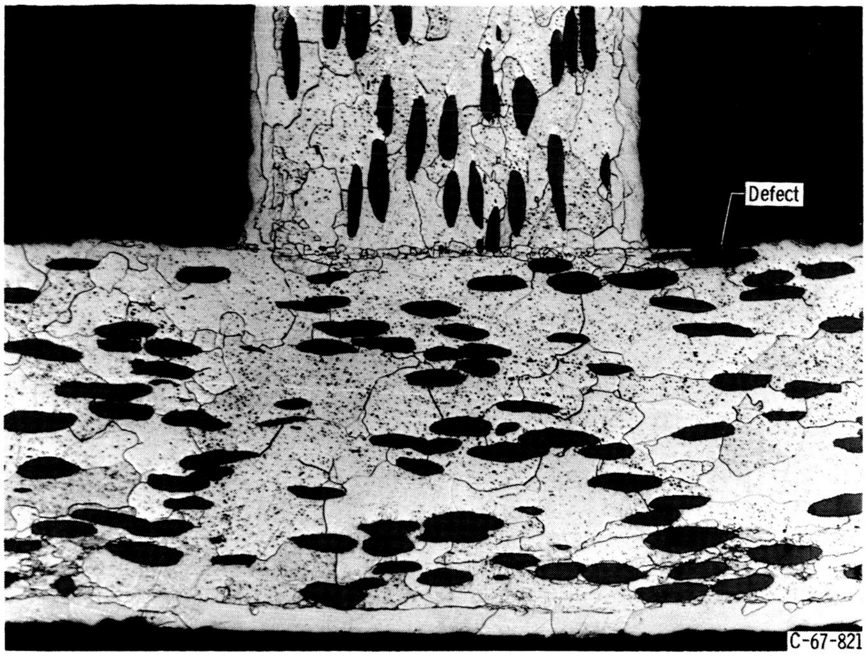

Figure 11. - Cross section of gas-pressure bonded T-joint specimen. Etchant, Murakami's reagent. Magnification, X100.

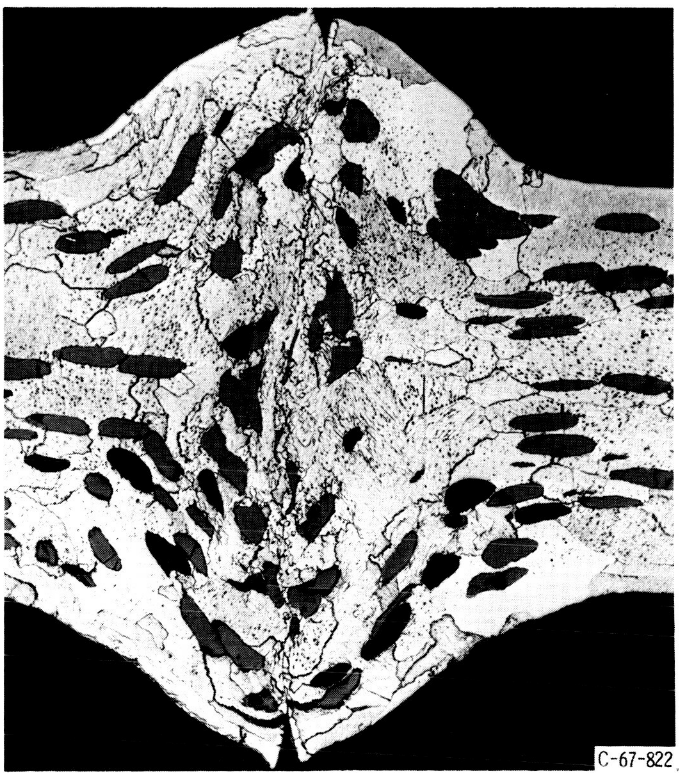

Figure 12. - Cross section of magnetic force welded small butt joint in tungsten - uranium dioxide. Etchant, Murakami's reagent. Magnification, X100.

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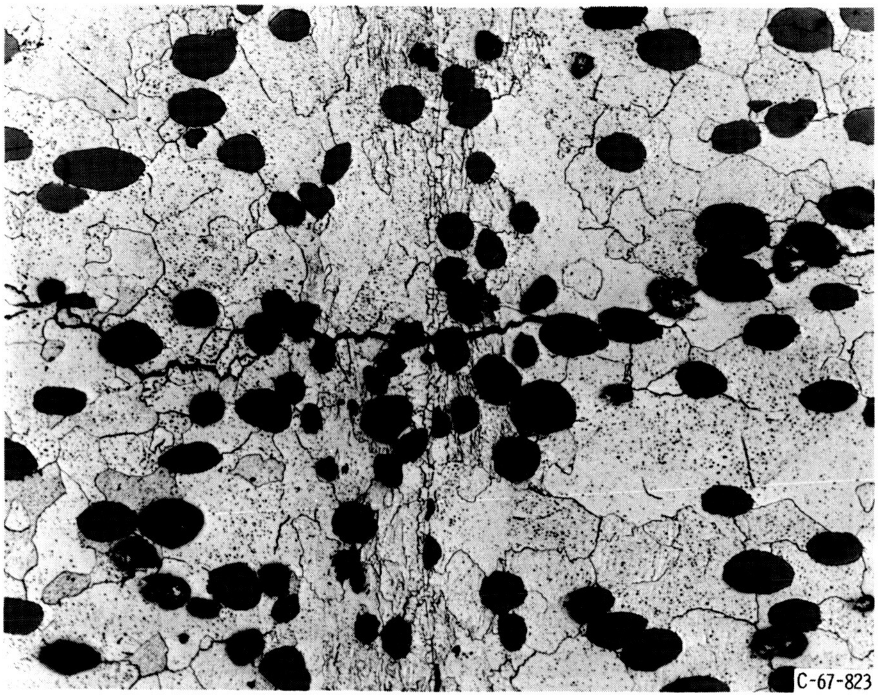

Weld interface

Figure 13. - Magnetic force welded tungsten - uranium dioxide plate sectioned parallel to major faces to show transverse crack. Etchant, Murakami's reagent. Magnification, X100.

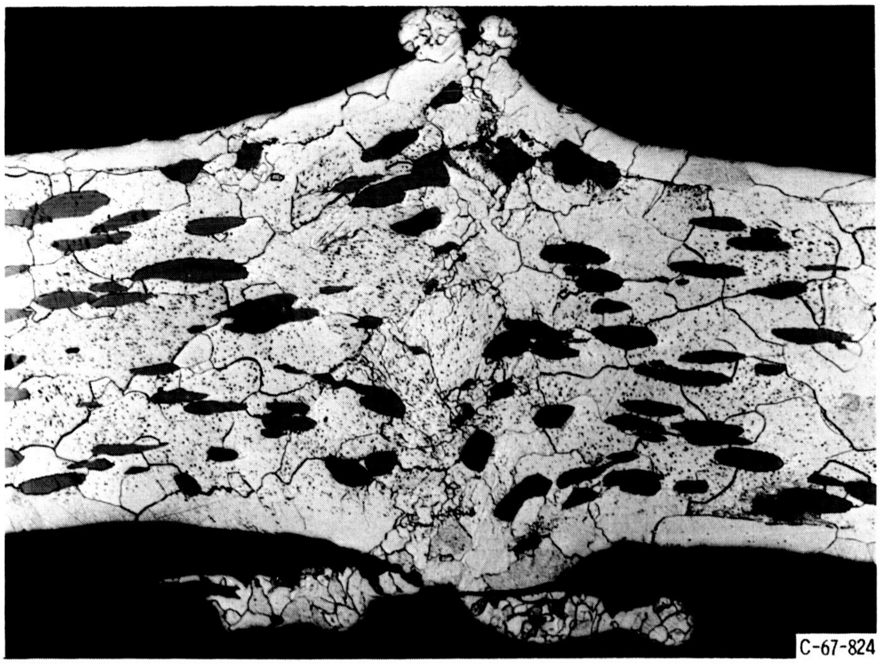

Figure 14. - Cross section of magnetic force welded tungsten - uranium dioxide cylinder. Etchant, Murakami's reagent. Magnification, X100.

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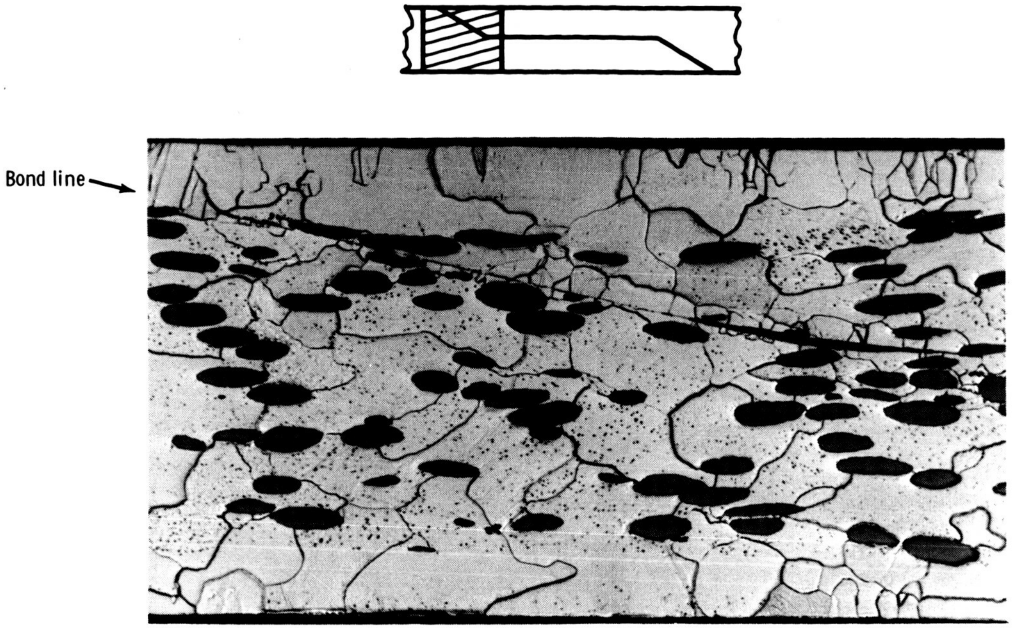
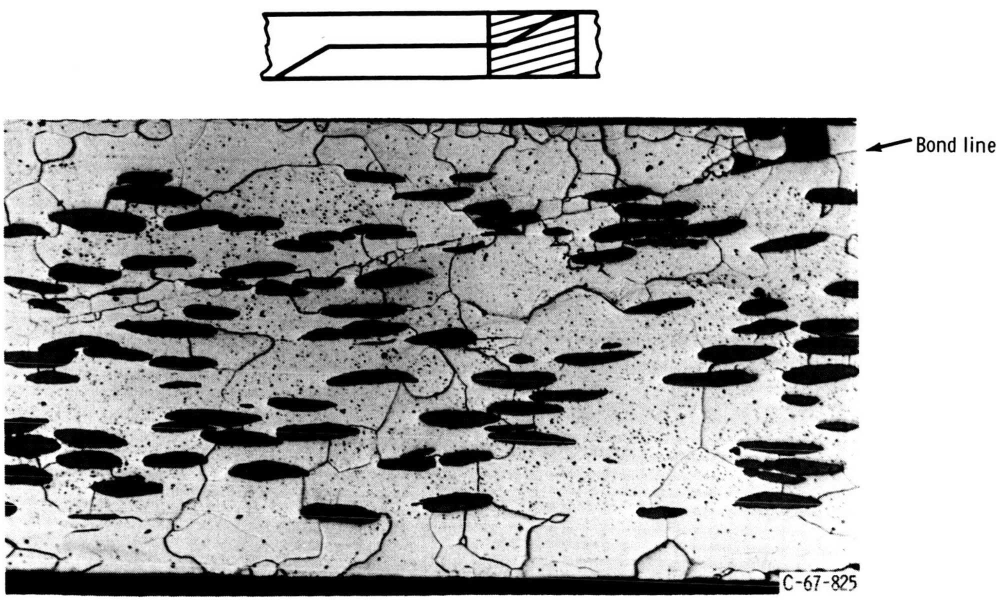


(a) Specimen 39. Joint classification, poor. Nonbonded region due to insufficient plastic flow.



(b) Specimen 44. Joint classification, fair. Note edge crumbling.

Figure 15. - Cross section from vacuum hot-press bonded small butt joints at location indicated. Etchant, Murakami's reagent. Magnification, X100.

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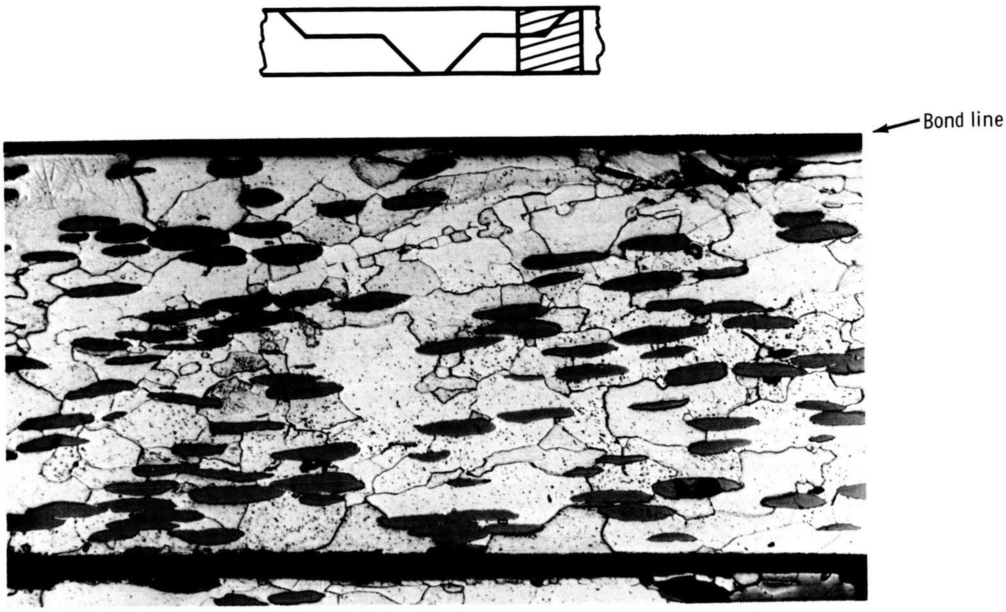
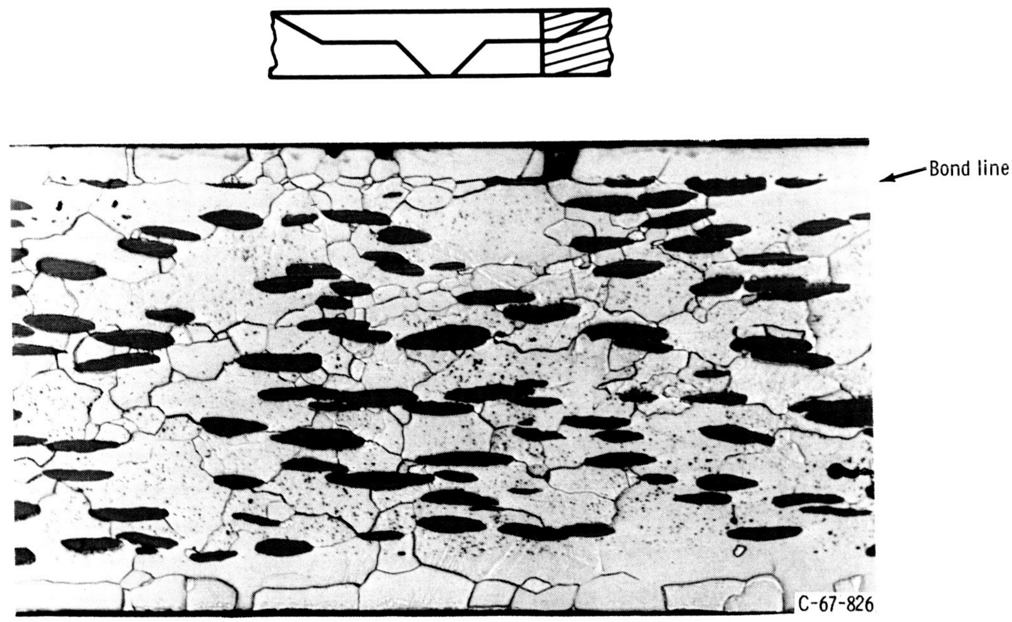


(c) Specimen 51A, insert-type joint. Joint classification, good.



(d) Specimen 48, insert-type joint. Joint classification, excellent.

Figure 15. - Concluded.

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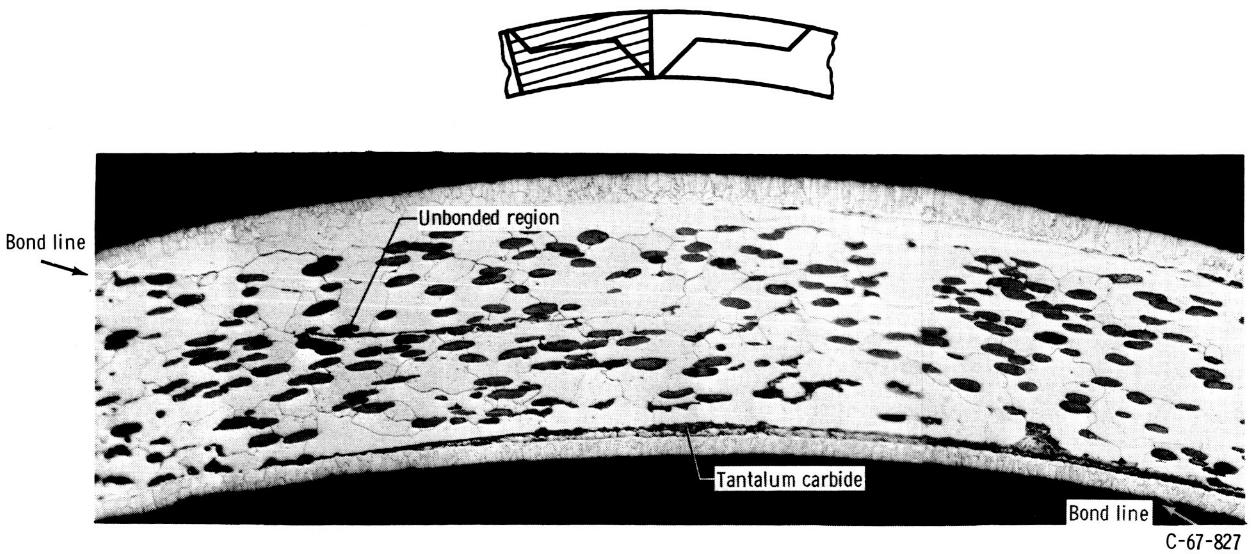

Figure 16. - Cross section of vacuum hot-press bonded cylinder butt joint at location indicated. Specimen, 0; joint classification, good; etchant, Murakami's reagent. A layer of foreign material exists on inside diameter between the composite and vapor deposited tungsten. The foreign material is believed to be tantalum carbide. Magnification, X50.

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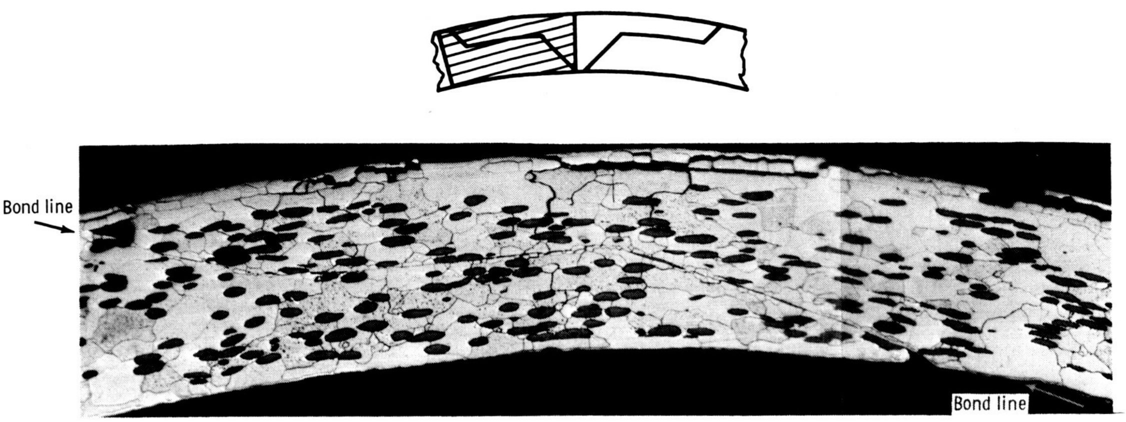
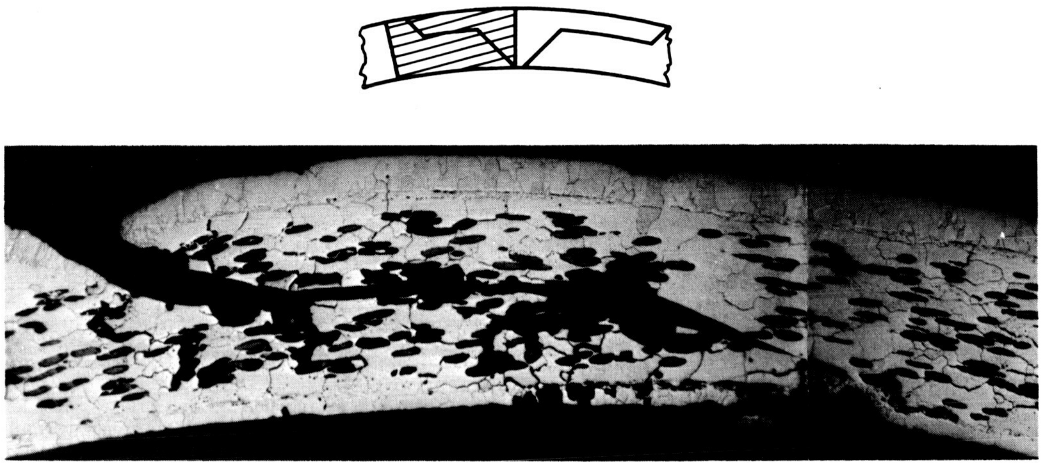
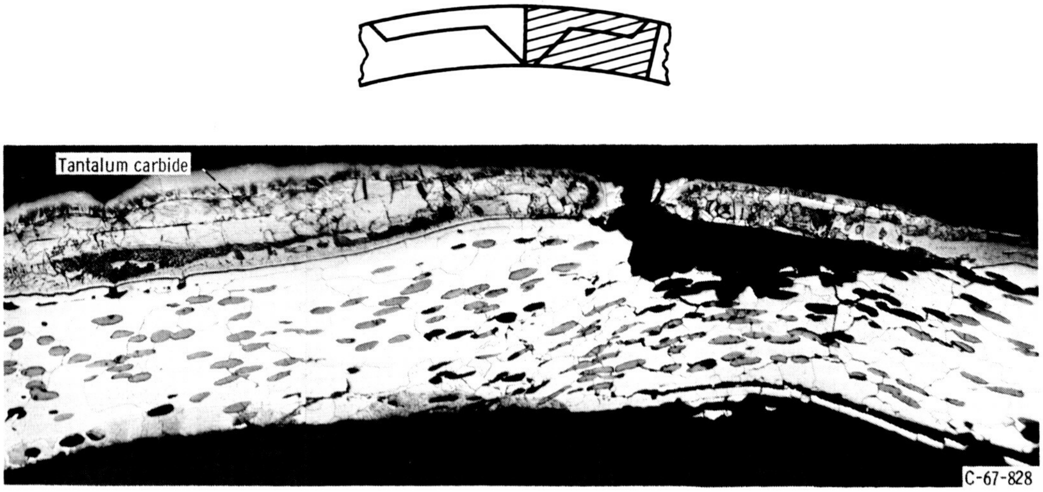


(a) Cross section of cylinder butt joint (specimen F) with little grain growth across interface. Joint classification, poor.



(b) Cross section of cylinder butt joint (specimen I) showing separated joint and uranium dioxide loss at interface.



(c) Cross section of cylinder butt joint (specimen R) showing excessive plastic flow and loss of material at joint. Tantalum carbide contamination on outside diameter.

Figure 17. - Poorly bonded and defective joints in tungsten - uranium dioxide cylinders joined by vacuum hot-pressing. Joints from locations indicated. Etchant, Murakami's reagent. Magnification, X50.

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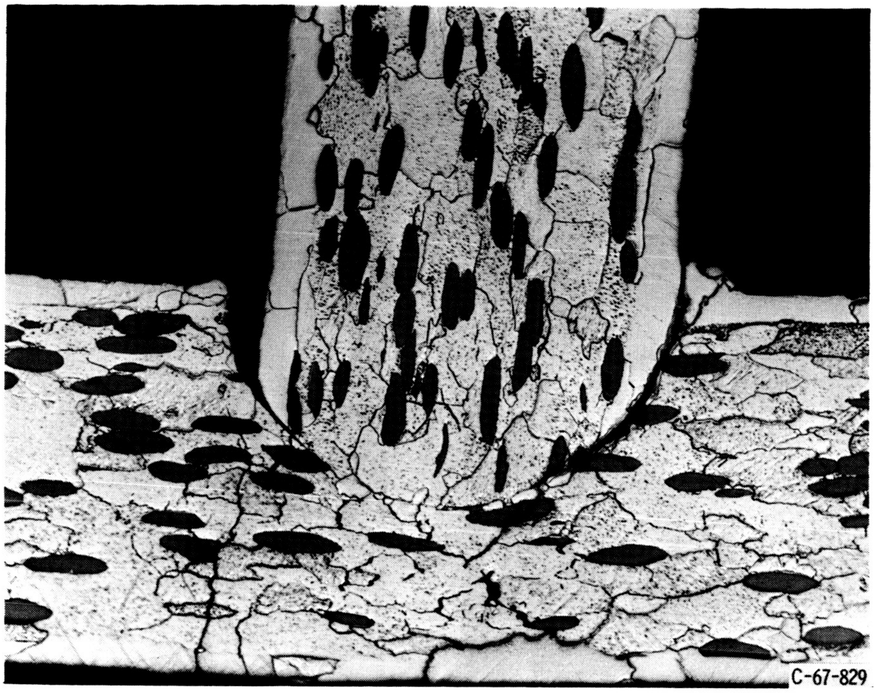

Figure 18. - Vacuum hot-press bonded T-joint showing large unbonded areas. Extensive cracking below vertical member. Etchant, Murakami's reagent. Magnification, X100.

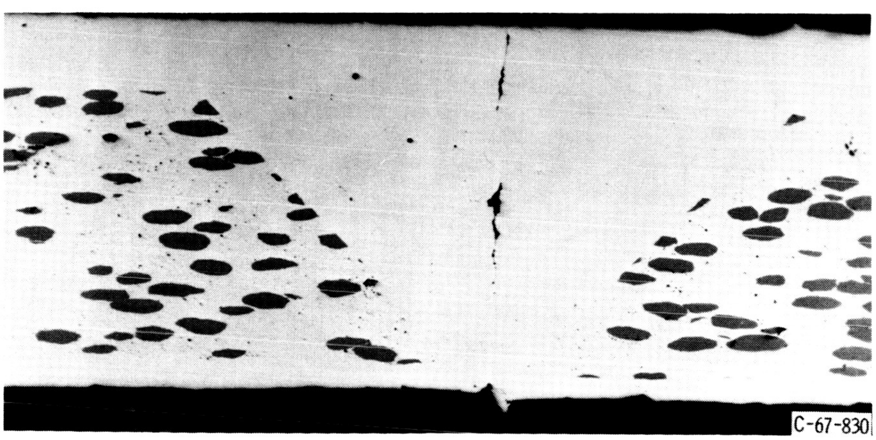

Figure 19. - Typical centerline void in vapor deposition welded joint. Joint heat treated after welding, at 1925° C; unetched. Magnification, X75.

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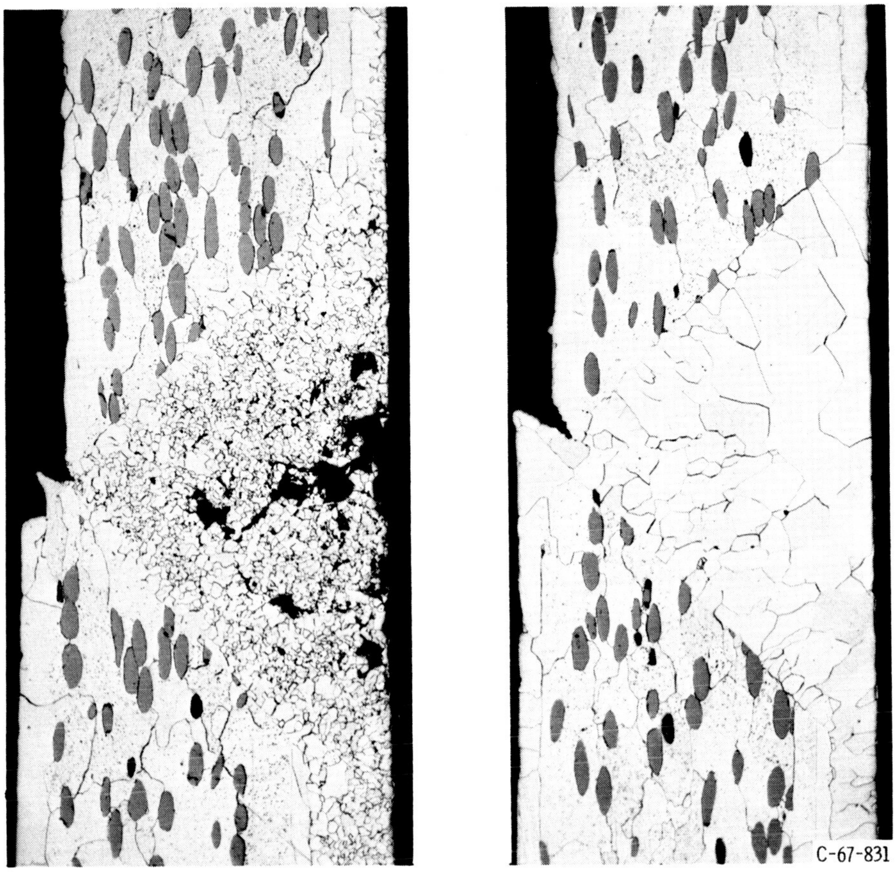

(a) Midlength of joint. (b) 1/8-Inch from one end.

Figure 20. - Cross section of vapor deposition welded joint at two different locations showing both defective fine grained region and high integrity welded region. Joint heat treated after welding at 1925° C; etchant, Murakami's reagent. Magnification, X75.

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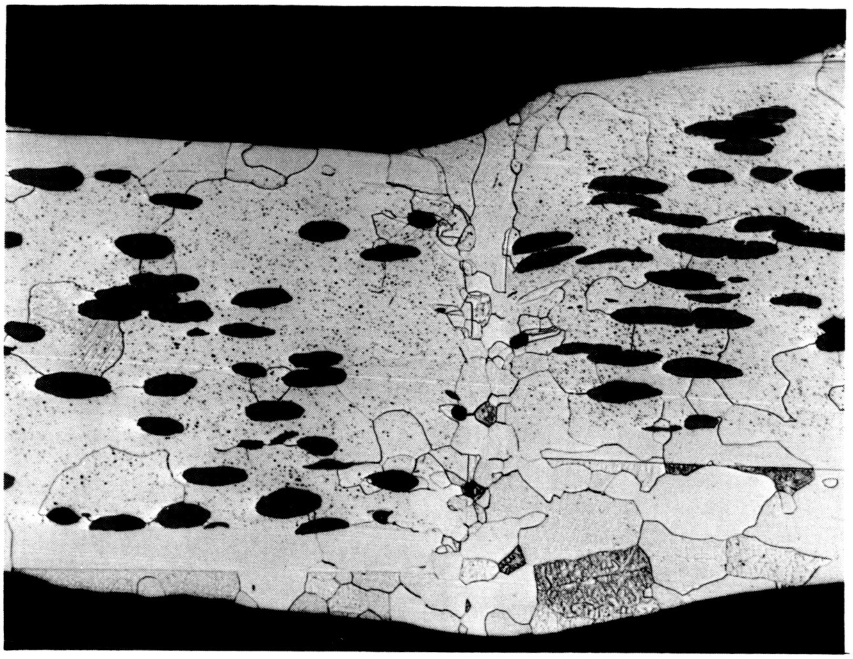
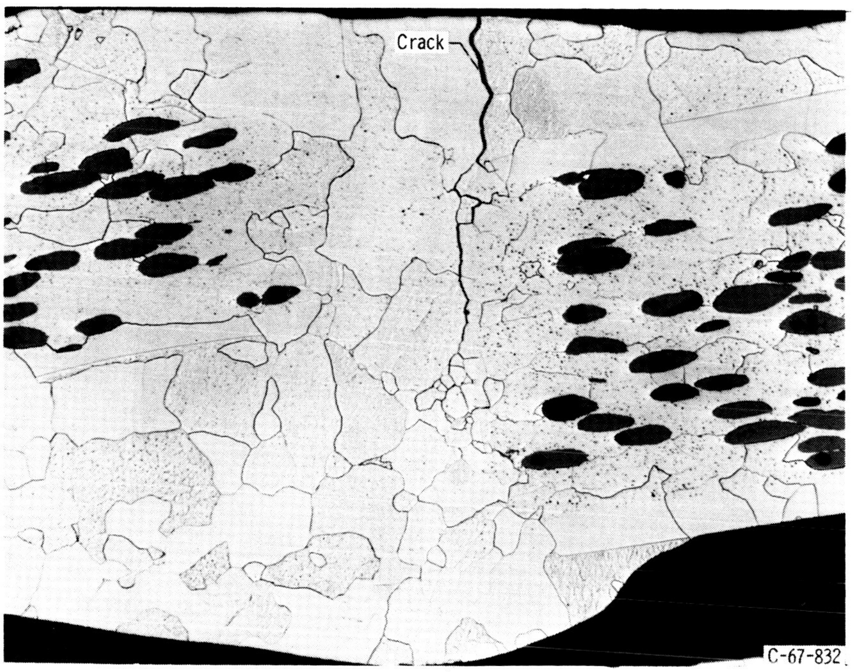


Joint

(a) Sound joint specimen 276; but poor alinement.



Joint

(b) Joint 289 illustrating intergranular cracking. Poor joint alinement and base material dissolution are also evident.

Figure 21. - Cross sections of small butt joints brazed with the tungsten - 50-percent molybdenum - 3-percent rhenium alloy. Voids within uranium dioxide particles and loss of entire particles are believed to have developed during metallographic preparation. Etchant, Murakami's reagent. Magnification, X100.

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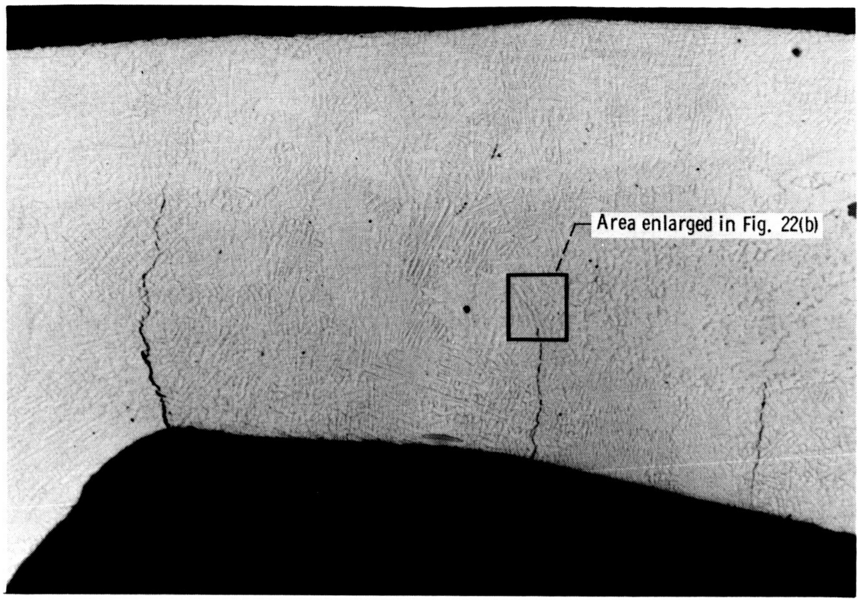
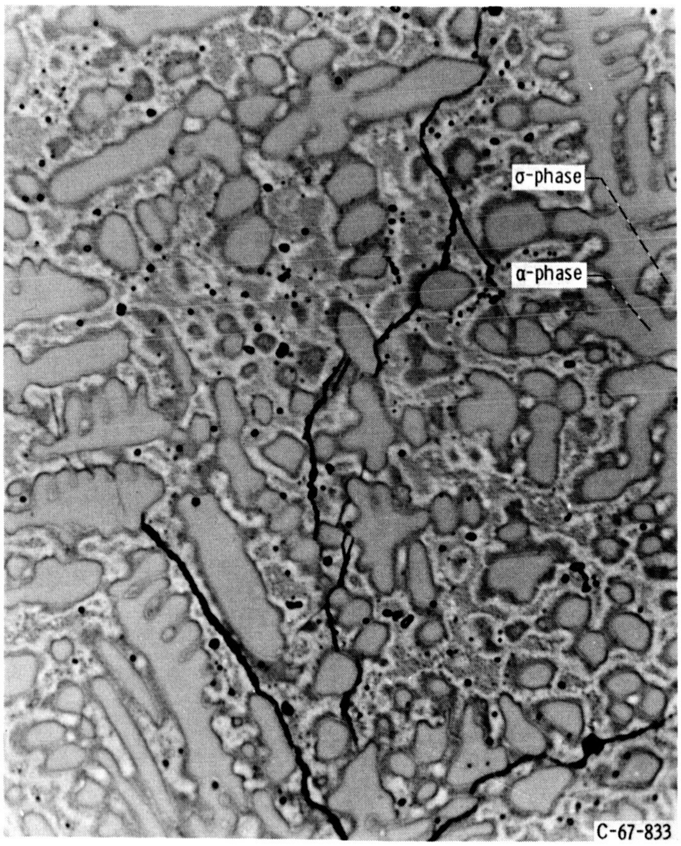
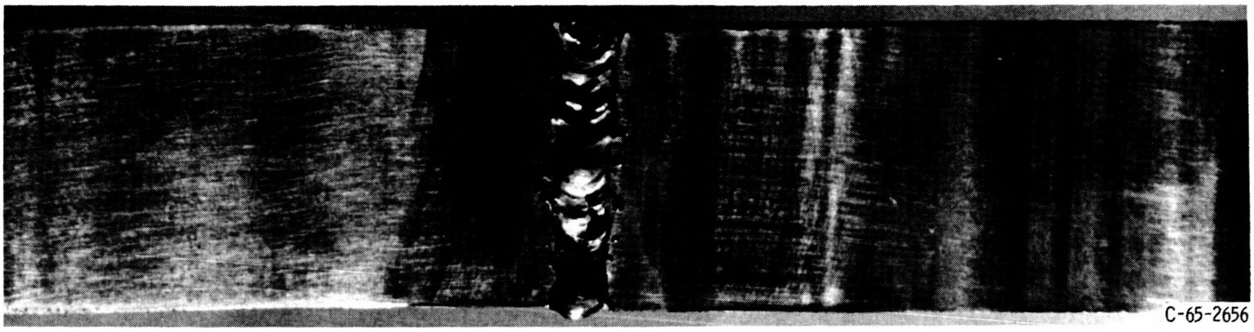


(a) Low magnification. (X100). Unetched.



(b) High magnification. (X1000). Etchant, Murakami's reagent.

Figure 22. - Longitudinal section through joint gas tungsten-arc brazed with tungsten - 25-percent osmium illustrating braze metal cracking transverse to joint. Specimen 304.



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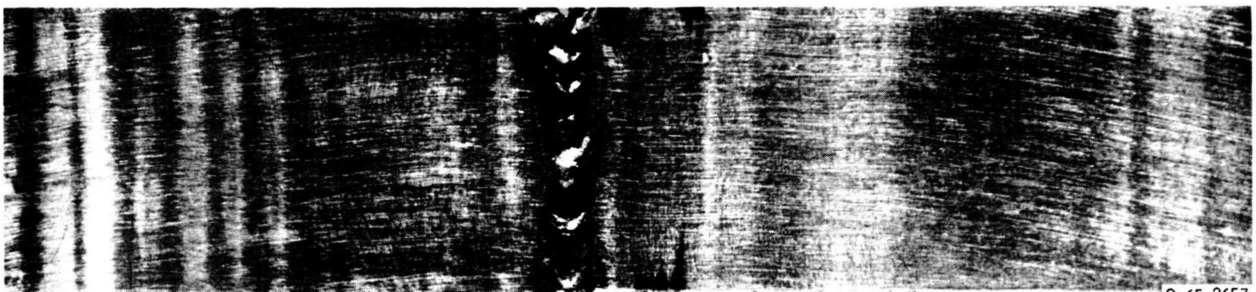
Face (top)



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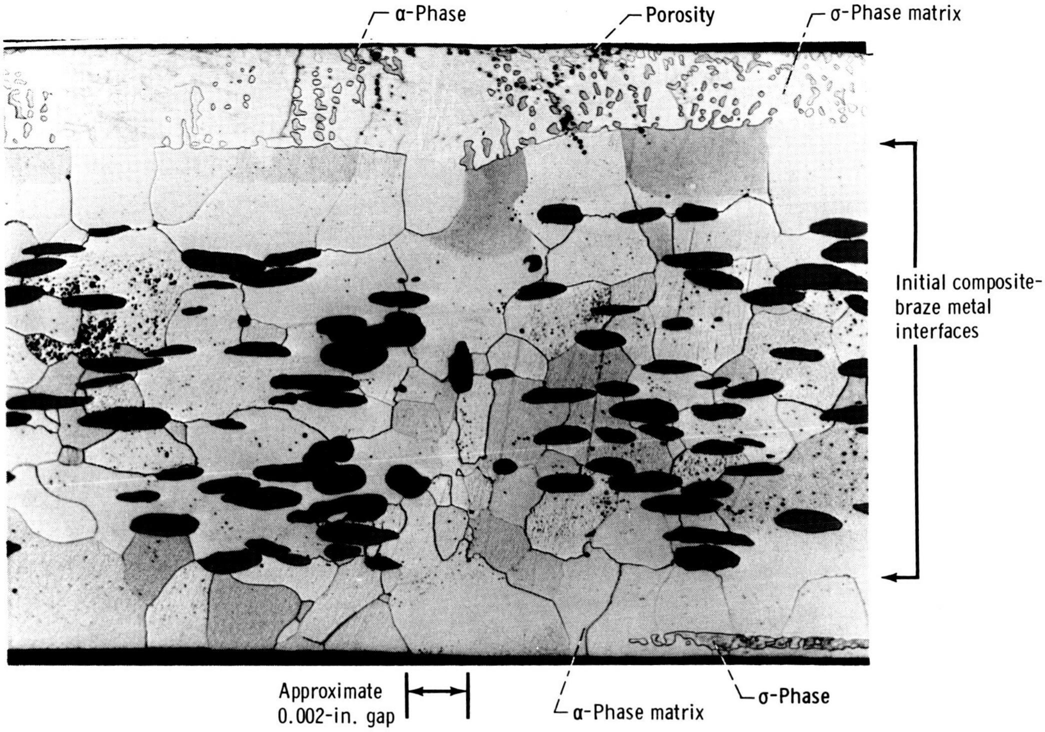
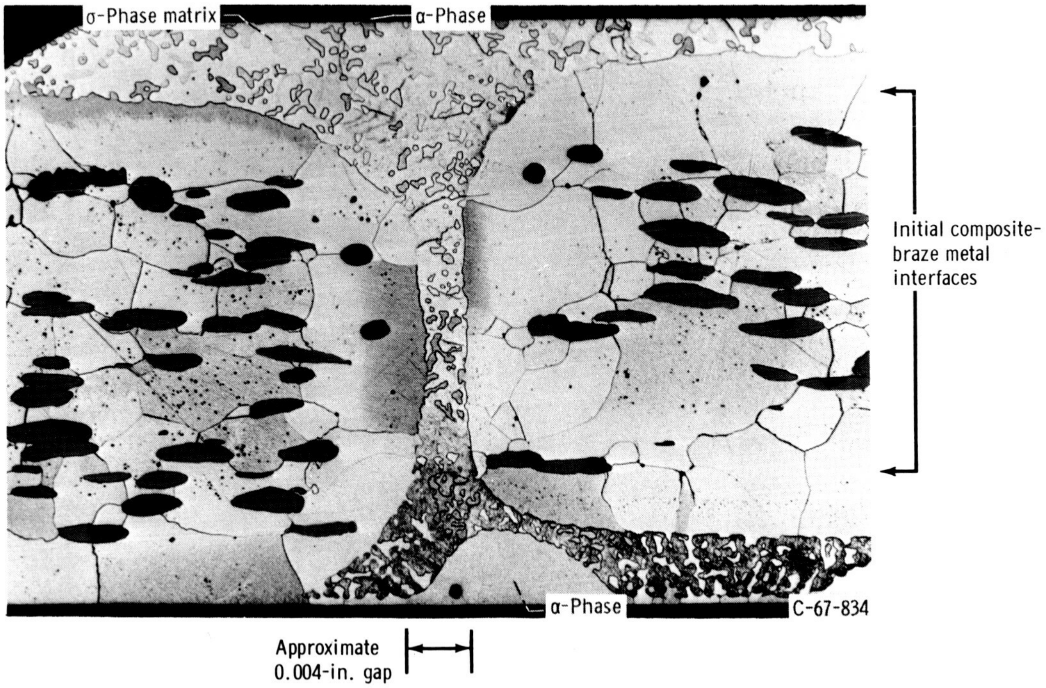
Root (bottom)

Figure 23. - Tensile blank specimen 356 gas tungsten-arc brazed with tungsten - 25-percent osmium alloy. Arc-start and runoff tabs (shown in fig. 6) have been removed.



(a) Section 3/16-inch on one side of midlength of joint.



(b) Section 3/16-inch on other side of midlength of joint.

Figure 24. - Cross sections of tungsten - 25-percent osmium braze joint in the tungsten - uranium dioxide composite after heating for 10 hours at 2500° C in hydrogen. The loss of uranium dioxide (black areas) is believed to have occurred during metallographic preparation. Etchant, Murakami's reagent. Magnification, X100.

Specimen 365C: tungsten - 25 percent osmium

Specimen 370C: molybdenum - 5 percent osmium

Specimen 372C: tungsten - 50 percent molybdenum - 3 percent rhenium

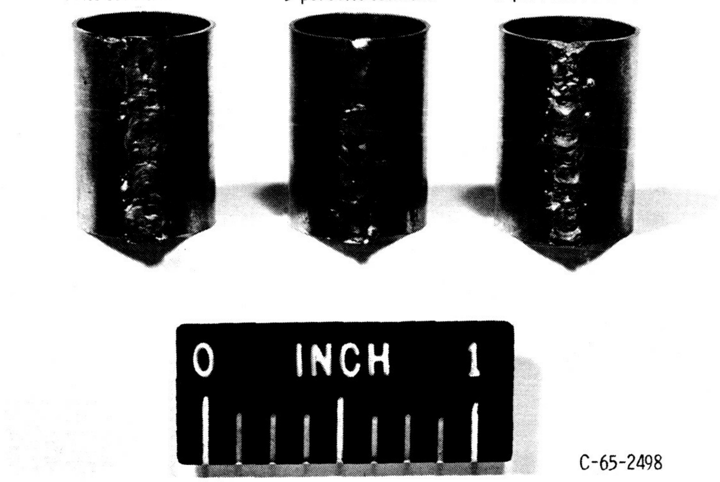


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Figure 25. - Tungsten - uranium dioxide cylinders joined by gas tungsten-arc brazing by using three different braze alloys.

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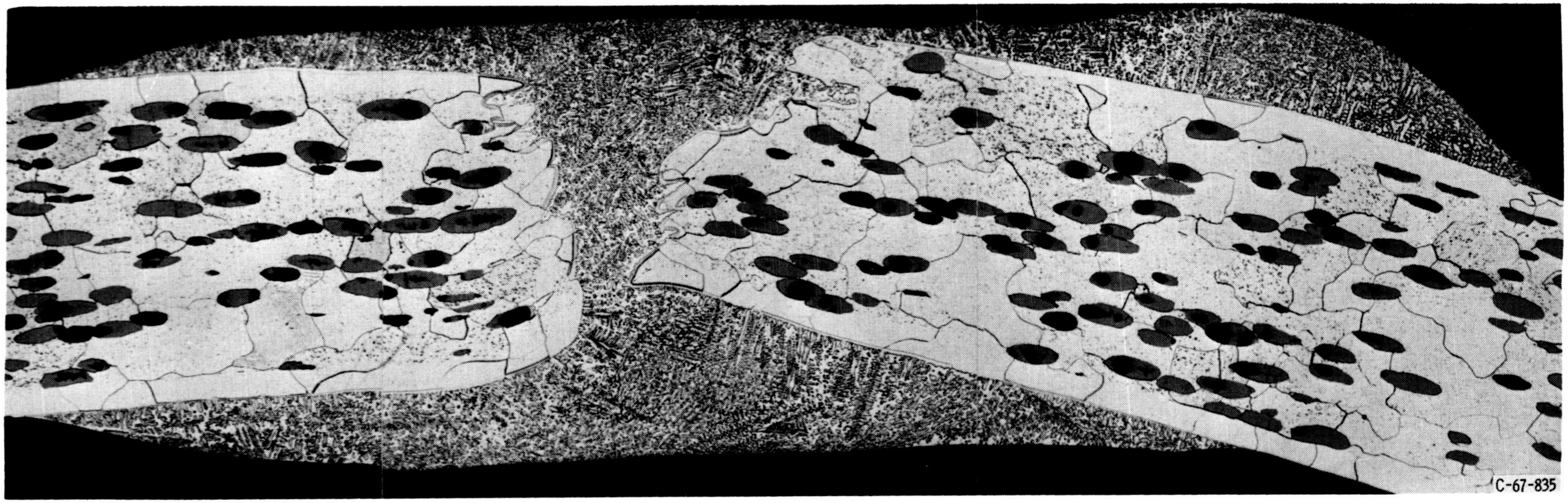

Figure 26. - Typical cross section of as-brazed joint in tungsten - uranium dioxide cylinders of tungsten - 25-percent osmium alloy. The loss of uranium dioxide (black areas) is believed to have occurred during metallographic preparation. Etchant, Murakami's reagent. (Reduced 15 percent in printing.) Magnification, X100.

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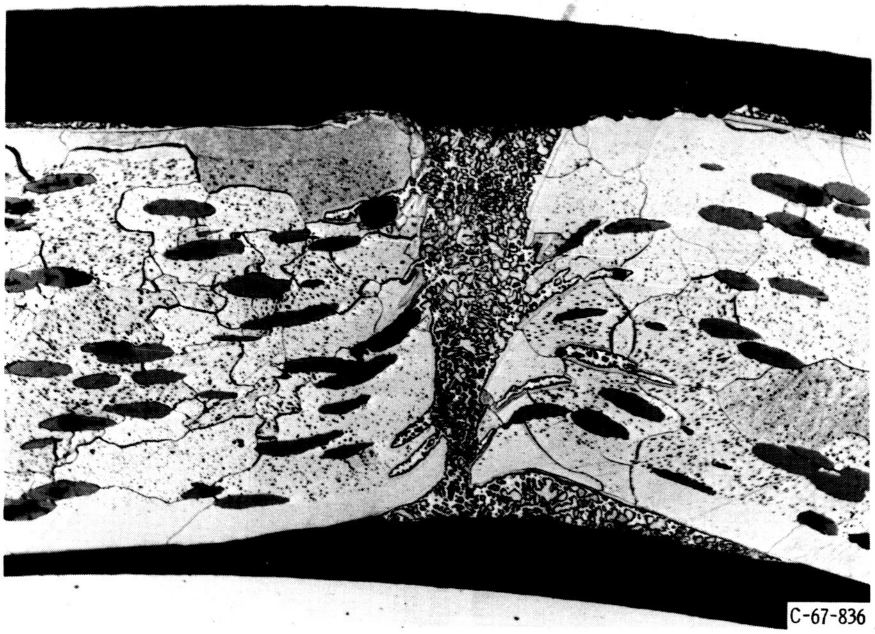
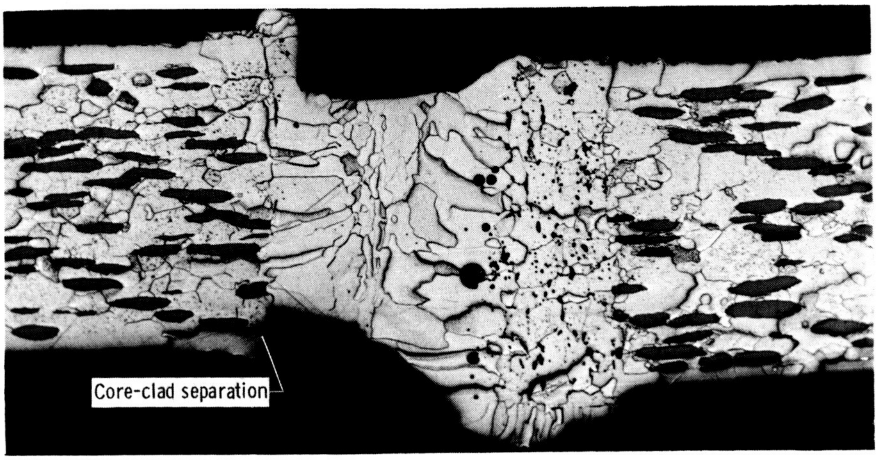
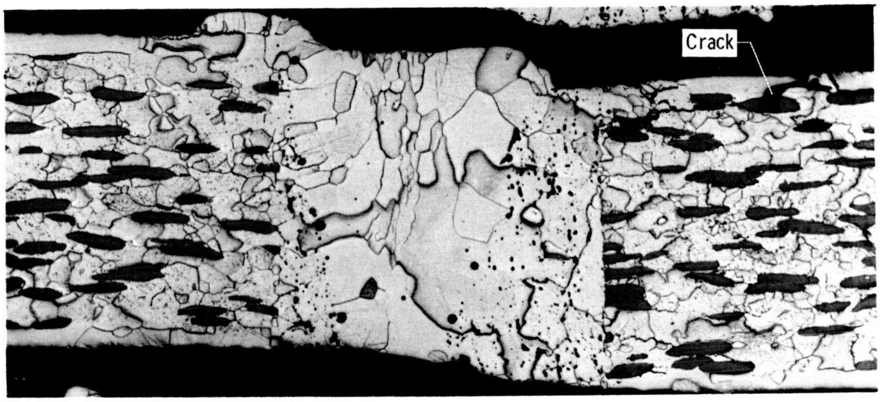


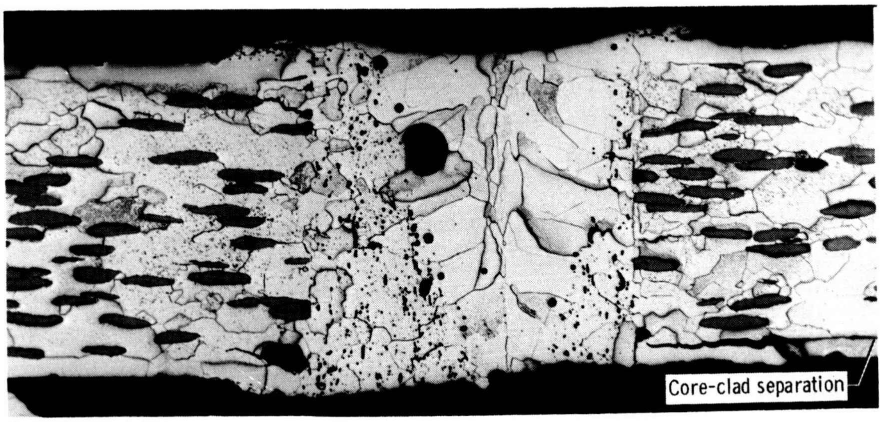
Figure 27. - Cross section of excellent as-brazed joint in tungsten-uranium dioxide cylinder using tungsten - 25-percent osmium alloy. Loss of uranium dioxide (black areas) is believed to have occurred during metallographic preparation. Etchant, Murakami's reagent. Magnification, X100.



(a) Near start.



(b) Midlength.



C-67-837

(c) Near finish.

Figure 28. - Butt joint in tungsten-uranium dioxide composite produced by special electron beam welding technique. Etchant, Murakami's reagent. Magnification, X75.

[redacted]

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